

STOCHASTIC ANALYSIS OF PARTICULATE PROCESSES - A STUDY OF ATTRITION,  
SIEVING AND GRINDING

by

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A MASTER'S THESIS

submitted in partial fulfillment of the  
requirements for the degree

MASTER OF SCIENCE

Department of Chemical Engineering

KANSAS STATE UNIVERSITY

Manhattan, Kansas

1986

Approved by:



Major Professor

## ACKNOWLEDGEMENTS

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The author wishes to express his gratitude and sincere thanks to his major professor, Dr. L. T. Fan, University Distinguished Professor and Head of Chemical Engineering Department at Kansas State University for his continuous guidance throughout the preparation of this thesis. This thesis would have been, indeed, far inferior to its present state without his high standards in manuscript preparation and attention to detail. Thanks are also due to the committee members, Dr. R. Nassar, Professor of Statistics, Dr. W. P. Walawender, Professor of Chemical Engineering and Dr. L. E. Erickson, Professor of Chemical Engineering, Kansas State University.

The author also wishes to express his appreciation to Mr. Rodney Fox for the suggestions with regard to the formulation of the master equation and to the other members of the stochastic research group for their suggestions.

The research assistantship awarded by the Engineering Experiment Station, Kansas State University through the Department of Chemical Engineering, for the academic years 1984-1986 is gratefully acknowledged.

The author dedicates this work to his parents Mr. Rajabapa Rao and Mrs. Vani Duggirala.

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## CHAPTER 1

### INTRODUCTION

#### THE MASTER EQUATION IN THE DESCRIPTION OF PARTICULATE PROCESSES

M. E. Harr (1977) states:

"At first glance, the probabilistic approach will seem like a poor, even overly sophisticated substitute for Newtonian determinism, but subsequent development will reveal the striking fact that deterministic solutions are nothing but special approximations of probabilistic models"

A process or system has been traditionally analyzed based on a continuum approach disregarding the existence of discrete elements in the material system, e.g., particles, bubbles, droplets, or molecules. In this approach the bulk properties of the system are obtained by simply averaging over a large number of these elements. In a particulate system or process, the particulate phase consists of definitely distinct discrete elements neither macroscopic nor microscopic in size; in other words, these elements are mesoscopic in size. The system contains a finite and often a relatively small number of particles, and the size and shape of the particles often vary extensively. These characteristics negate any justification for analyzing and modeling the system by resorting to the continuum approach. More often than not a particulate system or process has been observed to exhibit fluctuations in its behavior around that corresponding to its bulk properties (see, e.g., Kapur et al., 1977).

In the light of an increasingly important role played by particulate processes in the industry, a systematic or unified approach to analyze and describe them is urgently needed; such an approach has not been available hitherto.

The complete description of a physical or chemical process would require a knowledge of its microscopic state and behavior as governed by the fundamental laws of mechanics (see, e.g., Van Kampen, 1962). For a particulate process or system, this is tantamount to a description of the physical characteristics of an individual particle and its location coordinates in the system, its motion utilizing the familiar differential equations of mechanics based on Newton's laws, and the most formidable, the changes in the physical characteristics of the particle (for e.g., in grinding, the deformation of a particle brought about by the interplay of various stress distributions within the particle).

Obviously, models of a particulate process based on the microscopic equations are by no means simple to formulate and solve. On the other hand, the process can be described in an approximate and incomplete fashion by a relatively few "macroscopic variables". These variables are self-contained in the sense that they obey the phenomenological equations, which are differential equations of a deterministic type, i.e., the future values are determined from the initial ones; the changes in the state take place according to a specific rate mechanism (van Kampen, 1962).

Given the description of the microscopic, classical statistical mechanics determines the macroscopic behavior that can be expected using elements of probability theory. The employment of the tools of

classical statistical mechanics has yielded useful results only in the study of systems at equilibrium and comprising a very large number of entities ( $\sim 10^{23}$ ) (see, e.g., Mayer and Mayer, 1940 and Huang, 1963). For systems far from equilibrium or in which finite numbers of entities exist, an alternate approach is therefore required.

"Dynamic" stochastic theory provides probabilistic techniques for calculations based on a mesoscopic approach that resides in between the macroscopic and microscopic descriptions - the mesoscopic approach is more detailed than the macroscopic description, as it includes fluctuations but is withdrawn from the rigor of the formulation and solution of microscopic equations by means of the repeated randomness assumption - a manifestation of the Markov assumption. One such technique at an intermediate level between the microscopic and macroscopic equations is the master equation, an equation for the probability distribution of the various possible states of the system, where its evolution is described as a stochastic process.

The master equation describes the time dependence of the probability distribution of a set of dynamic variables of the system in continuous and discrete state space in an extremely simple form, in contrast to the microscopic equations. In the case of a multivariate master equation, in addition to the variances and covariances which allow for an extended description of the system beyond the deterministic mean value equations, the derivation of correlation functions for the variables involved enables the determination of rate constants from steady state data. Multivariate processes are encountered frequently in



chemical engineering, for instance, chemical reaction involving more than one species, particulate processes involving particle size distributions and in general, transport processes in phase space, such as absorption-desorption problems. In simple cases, explicit analytical solutions to the master equations exist. In general, however, such solutions are extremely difficult, if not impossible. This is especially so when the behavior of non-linear systems is described. For such systems, a systematic or rational approximation procedure can be adopted to obtain the desired results. One such procedure is the so-called system size expansion (see, e.g., Van Kampen, 1976). This procedure also provides the answer to the question of how the deterministic macroscopic equation emerges from the stochastic description in terms of a master equation and allows for the expression of the changes in the system behavior in terms of intensive variables, ameliorating scale-up and applicability. Such an approximation procedure still yields a greater depth of insight into the system behavior and is simpler than the formulation and solution of the microscopic equations.

There are essentially two conceptual approaches to a stochastic formulation of the system behavior and the establishment of a corresponding master equation. In one approach, we pose, in an ad hoc manner, rate problems in terms of probability theory (stochastic theory of birth and death processes, for instance) on the basis of intuitive perception about the nature and mechanisms of the rate process without reference to deterministic dynamics. The results are validated by experimental observations. Examples of such a formulation can be found

in numerous works in chemistry, genetics, epidemiology and ecology. In the other approach, we consider the stochastic formulation as an improvement over a deterministic formulation. For example, in analyzing a chemical reactor, the stochastic theory presumably takes into account the concentration fluctuations of reactants and products in small subvolumes of the overall reactor volume and thus yields information on the deviations of the reacting species about their deterministic mean (Oppenheim et al., 1977).

A particulate process or system lends itself to a stochastic analysis due to the finite number of mesoscopic entities present in volume elements, random behavior of the individual particles and the complexities encountered in attempts to analyze on a particle scale (particle motion and changes in physical characteristics). In this study the first approach for formulating stochastic models described above is employed to study the different particulate processes, chemical attrition in fluidized beds, sieving operations, and comminution. The master equation is formulated for the sieving and grinding processes. The system size expansion method, one of the approximation procedures is used to obtain a solution to the master equations that are otherwise not amenable to explicit solutions. A model based on the well known Migration and Illness-Death stochastic processes has been derived for describing chemical attrition in fluidized beds and grinding by abrasion. The applications of these models are illustrated through numerical simulations. The fluctuations which can be expected in such processes are also described. These yield valuable process

characteristics important to the study and progress of process development.

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## CHAPTER 2

### STOCHASTIC ANALYSIS OF ATTRITION - A GENERAL CELL MODEL

#### INTRODUCTION

Attrition is frequently encountered in various process operations such as fluidization, comminution and solids transport, either as a desired feature or as an undesired behavior. In all such processes the changes in the particle size distributions are random in nature (see, e.g., Wei et al., 1977, and Fan and Srivatsava, 1981); thus a stochastic treatment yields valuable information on the fluctuations arising thereof.

Attrition here denotes the size reduction of particles through surface removal or abrasion leaving the particles in adjacent smaller size intervals, where the particle size distribution is approximated as a series of particle size intervals, as in practice. The particle behavior, especially the transient evolution of the particle size distribution (e.g., bed loss during start up in a fluidized bed process), influences significantly the product characteristics, operability, economics, etc.

In this work a general attrition model is derived, which is sufficiently flexible and applicable to various processes. The application of the model to attrition in fluidized beds in the presence of chemical reaction, namely, chemical attrition, and to size reduction by abrasion is described. A numerical example of chemical attrition in a fluidized bed is provided to illustrate the application.

## MODEL DESCRIPTION AND DERIVATION

The system modeled here comprises a cell coupled to multiple inlets and outlets (see Figure 1). The particle sizes are distributed through  $s+1$  size ranges or intervals, or simply  $s+1$  sizes, beginning with the largest size (based on particle diameter or equivalent) and ending with the  $(s+1)$ -th sink interval or fines consisting of all particles below a certain size. Particle disintegration is assumed to occur only in the cell, and particles undergo attrition in the mode as shown in Figure 2. A particle in the  $i$ -th size range is reduced to the  $(i+1)$ -th size range at any instant and simultaneously generates a number of fines or particles belonging to the sink interval; the number produced is related to the mass difference between the sizes involved.

### System Description in the Absence of Solids Inflow

Consider a particle population existing in  $s + 1$  states or size ranges in the cell; the inflow of particles to the cell is disregarded here. A particle in the size range  $i$  is reduced through attrition into that in size range  $j$  where  $j = i+1, i+2, \dots, s+1$ , remains in the same size range, or exits from the cell into any of the  $r$  outlet streams. Transitions between consecutive size intervals are governed by intensities of transition defined as follows:

$$v_{ij}\Delta t + o(\Delta t) = \text{probability that a particle in size interval}$$

$i$ , or of size  $i$ , at time  $t$  will be of size  $j$   
at time  $t+\Delta t$  for  $j=i+1$  for all  $i=1,2,\dots,s$   
within the cell,

$\mu_{ik}\Delta t + o(\Delta t)$  = probability that a particle of size  $i$  within

the cell at time  $t$  will exit into the  $k$ -th

outlet stream at time  $t+\Delta t$  for all

$i=1,2,\dots,s+1$  and  $k=1,2,\dots,r$

$o(\Delta t)$  is a function of  $\Delta t$  such that  $\lim_{\Delta t \rightarrow 0} \frac{o(\Delta t)}{\Delta t}$ , the inclusion of this

term implies that the probability of more than a single transition or

other events occurring in the time interval  $\Delta t$  is  $o(\Delta t)$ . Although the

derivation can be modified to accommodate a time-dependency of the tran-

sition intensities (i.e.,  $v_{ij}$  and  $\mu_{jk}$  as functions of time  $t$ ), they are

assumed to be independent of time  $t$ , i.e., the process is assumed to be

time homogeneous. Also, the probability that a particle in size inter-

val  $i$  at time  $t$  will remain in the same size interval within the cell at

time  $t+\Delta t$  for all  $i=1,2,\dots,s+1$ , is  $1+v_{ii}\Delta t+o(\Delta t)$ , where  $v_{ii}$  is defined

as

$$v_{ii} = - \left[ v_{i,i+1} + \sum_{k=1}^r \mu_{ik} \right], \text{ for all } i=1,2,\dots,s$$

satisfying the condition that the sum of the probabilities of transition

from state  $i$  to all other states including itself is unity, and

$$v_{s+1,s+1} = - \sum_{k=1}^r \mu_{s+1,k}$$

Note that subscripts  $i$  or  $j$  refers to any of states  $1,2,\dots,s+1$  within

the cell and subscript  $k$  refers to any of outlet streams  $1,2,\dots,r$ . The

intensity  $v_{ii} < 0$  for all  $i=1,2,\dots,s+1$ ; in the case of a batch process,

for which the outflow is also absent, we have  $\mu_{ik}=0$  for all  $k=1,2,\dots,r$ ,

and  $v_{s+1,s+1} = 0$  since the sink interval is an absorbing state (see, e.g., Chiang, 1980). The transition probabilities are defined in the following manner.

$p_{ij}(\tau, t)$  = probability that a particle of size  $i$  at time  $\tau$  will be that of size  $j$  at time  $t(t > \tau)$  within the cell for all  $i, j = 1, 2, \dots, s+1$ ,

$q_{ijk}(\tau, t)$  = probability that a particle of size  $i$  at time  $\tau$  within the cell will be that of size  $j$  when exiting from the cell through outlet stream  $k$  by time  $t$  for all  $i, j = 1, 2, \dots, s+1$  and  $k = 1, 2, \dots, r$

The initial conditions satisfied by these probabilities are:

$$p_{ii}(\tau, \tau) = 1$$

and

$$p_{ij}(\tau, \tau) = 0, \quad i, j = 1, 2, \dots, s+1; \quad i \neq j$$

$$q_{ijk}(\tau, \tau) = 0, \quad i, j = 1, 2, \dots, s+1; \quad k = 1, 2, \dots, r.$$

In the case of attrition, the following conditions hold;

$$p_{ij}(\tau, t) = 0$$

and

$$q_{ijk}(\tau, t) = 0, \quad \text{for all } i > j \text{ and } t$$

Assuming that the process under consideration possesses a Markovian nature, explicit formulae for the transition probabilities can be derived based on the transition intensities (see APPENDIX A). Being



Markovian and time homogeneous, the transition probabilities of the process depend only on the time difference  $t-\tau$  and not on  $t$  or  $\tau$  separately; hence letting  $\tau=0$  and  $t$  represent the time interval length,

$$p_{ij}(0,t) = \sum_{m=1}^{s+1} \frac{A'_{ij}(\rho_m)}{\prod_{\substack{n=1 \\ n \neq m}}^{s+1} (\rho_m - \rho_n)} e^{\rho_m t} \quad (1a)$$

or

$$p_{ij}(0,t) = \sum_{m=1}^{s+1} \frac{A'_{ij}(\rho_m) T_{jm}(p)}{|T(p)|} e^{\rho_m t}, \quad (1b)$$

$$i=1,2,\dots,s+1 \text{ and } j=i+1,\dots,s+1$$

and

$$p_{ij}(0,t) = 0, \quad j < i$$

$\rho_m$ ,  $m = 1,2,\dots,s+1$  are the eigenvalues obtained as the roots of the characteristic equation  $|\rho I - V'| = 0$ , in which  $V'$  is the transpose of the matrix whose elements are the transition intensities  $v_{ij}$ ;  $A'_{ij}(\rho_m)$  is the  $(i,j)$ th cofactor of  $A'(\rho_m) = |\rho I - V'|$ . The matrix  $T(p)$  is formed from the eigen vectors  $T_1(p), T_2(p), \dots, T_{s+1}(p)$ ; each eigenvector contains elements of the corresponding columns of the adjoint matrix  $A(\rho_m) = (\rho_m I - V)$ . The exit transition probabilities  $q_{ijk}(0,t)$  are obtained from  $p_{ij}(0,t)$  (see, e.g., Chiang, 1980). A particle of size  $i$  at time

$t=0$  may enter outlet stream  $k$  either directly from size  $i$  or after being reduced to a smaller size  $j$  at any time  $\tau$  prior to  $t$ . Thus,

$$q_{ijk}(0,t) = \int_0^t p_{ij}(0,\tau) \mu_{jk} d\tau \quad (2)$$

This expression is integrated to yield

$$q_{ijk}(0,t) = \sum_{m=1}^{s+1} \frac{A'_{ij}(\rho_m)}{\prod_{\substack{n=1 \\ n \neq m}}^{s+1} (\rho_m - \rho_n)} \cdot \frac{e^{\rho_m t} - 1}{\rho_m} \mu_{jk} \quad (3a)$$

or

$$q_{ijk}(0,t) = \sum_{m=1}^{s+1} A_{pi}(\rho_m) \frac{T_{jm}(p)}{|T(p)|} \cdot \frac{e^{\rho_m t} - 1}{\rho_m} \mu_{jk}, \quad (3b)$$

$$i=1,2,\dots,s+1, \quad j=i,i+1,\dots,s+1 \quad \text{and} \quad k=1,2,\dots,r$$

and

$$q_{ijk}(0,t) = 0, \quad \text{for all } j < i$$

A particle may exist in any of the size ranges within the cell at any time  $t$ , or it may exit through any of the outlet streams by time  $t$ . For a particle of size  $i$  initially, therefore,

$$\sum_{j=i}^{s+1} p_{ij}(0,t) + \sum_{k=1}^r \sum_{j=i}^{s+1} q_{ijk}(0,t) = 1 \quad (4)$$

To obtain the particle size distribution in the system, it is necessary to define the following quantities.

$X_{ij}(t)$  = random variable representing the number of particles of size  $j$  in the cell at time  $t$ , which were of size  $i$  initially in the cell at time  $t=0$

$Y_{ijk}(t)$  = random variable representing the number of particles of size  $j$  exited from the cell through outlet stream  $k$  by time  $t$ , which were of size  $i$  initially in the cell at time  $t=0$

$X_j(t)$  = random variable representing the number of particles of size  $j$  in the cell at time  $t$  arising from the initial particle population

$Y_{jk}(t)$  = random variable representing the number of particles of size  $j$  exited from the cell through outlet stream  $k$  by time  $t$ .

Consider an initial particle size distribution  $\underline{m}(0)$ , i.e.,  $\underline{m}(0) = [m_i(0), i \in \{1, 2, \dots, s+1\}]$  where  $m_i(0)$  is the number of size  $i$  particles existing within the cell at time  $t=0$ . At any time  $t$ , these particles appear in any of the  $j$  size intervals within the cell or they exit through any of the  $k$  outlet streams by time  $t$ . Hence,

$$m_i(0) = \sum_{j=i}^{s+1} X_{ij}(t) + \sum_{k=1}^r \sum_{j=i}^{s+1} Y_{ijk}(t) \quad (5)$$

Assuming that each transition or outcome is independent of the other, i.e., the particles behave in a stochastically independent manner, we obtain a multinomial distribution given by

$$\text{Prob} [X_{1,1}=x_{1,1}, X_{1,1+1}=x_{1,1+1}, \dots, X_{1,s+1}=x_{1,s+1}; Y_{1,1,1}=y_{1,1,1}, Y_{1,1+1,1}=y_{1,1+1,1}, \dots, Y_{1,s+1,1}=y_{1,s+1,1}; Y_{1,1,2}=y_{1,1,2},$$

$$\begin{aligned}
& Y_{i,i+1,2} = y_{i,i+1,2}, \dots; Y_{i,s+1,2} = y_{i,s+1,2}; \\
& \dots; Y_{i,i,r} = y_{i,i,r}; Y_{i,i+1,r} = y_{i,i+1,r}, \quad Y_{i,s+1,r} = y_{i,s+1,r} \\
& = \frac{m_i(0)!}{\prod_{j=i}^{s+1} x_{ij} \prod_{k=1}^r \prod_{j=1}^{s+1} y_{ijk}} \prod_{j=i}^{s+1} (p_{ij}(0,t))^{x_{ij}} \prod_{k=1}^r \prod_{j=1}^{s+1} (q_{ijk}(0,t))^{y_{ijk}} \quad (6)
\end{aligned}$$

where the lower case letters of the random variables refer to the values they assume such that

$$m_i(0) = \sum_{j=i}^{s+1} x_{ij} + \sum_{k=1}^r \sum_{j=i}^{s+1} y_{ijk} \quad (7)$$

Here any symbol with a subscript 's+1' refers to the fines produced from the particles in a respective size range through successive 'core' attrition and not to the fines generated by attrition between consecutive size ranges.

The expected values and variances of various quantities and the covariances of pairs of these quantities can be obtained through binomial consideration (see, e.g., Mood et al., 1974):

$$E[X_{ij}(t)] = m_i(0)p_{ij}(0,t) \quad (8)$$

$$\text{Var}[X_{ij}(t)] = m_i(0)p_{ij}(0,t)(1-p_{ij}(0,t)) \quad (9)$$

$$E[Y_{ijk}(t)] = m_i(0)q_{ijk}(0,t) \quad (10)$$

$$\text{Var}[Y_{ijk}(t)] = m_i(0)q_{ijk}(0,t)(1-q_{ijk}(0,t)) \quad (11)$$

$$\text{Cov}[X_{ij}(t), X_{id}(t)] = -m_i(0)p_{ij}(0,t)p_{id}(0,t) \quad (12)$$

$$\text{Cov}[Y_{ijk}(t), Y_{idf}(t)] = -m_i(0)q_{ijk}(0,t)q_{idf}(0,t) \quad (13)$$

$$\text{Cov}[Y_{ijk}(t), Y_{ijf}(t)] = -m_i(0)q_{ijk}(0,t)q_{ijf}(0,t), \quad k \neq f \quad (14)$$

$$\text{Cov}[X_{ij}(t), Y_{idk}(t)] = -m_i(0)p_{ij}(0,t)q_{idk}(0,t) \quad (15)$$

$$i=1,2,\dots,s; j,d=i,i+1,\dots,s+1, j \neq d; k=1,2,\dots,r$$

and

$$E[X_j(t)] = \sum_{i=1}^j m_i(0)p_{ij}(0,t) \quad (16)$$

$$\text{Var}[X_j(t)] = \sum_{i=1}^j m_i(0)p_{ij}(0,t)(1-p_{ij}(0,t)) \quad (17)$$

$$E[Y_{jk}(t)] = \sum_{i=1}^j m_i(0)q_{ijk}(0,t) \quad (18)$$

$$\text{Var}[Y_{jk}(t)] = \sum_{i=1}^j m_i(0)q_{ijk}(0,t)(1-q_{ijk}(0,t)), \quad (19)$$

$$j=1,2,\dots,s$$

The fines present at any time  $t$ , arise from successive 'core' attrition of larger particles, initial amount of fines present in the cell or when particles of a certain size are reduced to the next size range. The amount of fines generated either from core attrition or as 'by product' by any time  $t$  is represented as  $X_{s+1}^{\text{gen}}(t)$  which is obtained through mass balance:

$$X_{s+1}^{\text{gen}}(t) = \left[ \sum_{i=1}^s m_i(0)w_i - \sum_{j=1}^s X_j(t)w_j - \sum_{k=1}^r \sum_{j=1}^s Y_{jk}(t)w_j \right] \quad (20)$$

where it is assumed that the particles in size range  $i$  have a mean weight of  $w_i$ . The accumulation of fines is cumulative. The amount of fines produced is retained as a quantity here and not a number. The number of fines can be obtained as the ratio of the amount to a

prescribed mean weight,  $w_{s+1}$ . It is possible to obtain the probability distribution of the fines conditional on the values of other quantities, but it is more useful to deduce the moments. The expected value and the variance of the amount of fines generated by time  $t$  are given by the following relations:

$$E[X_{s+1}^{\text{gen}}(t)] = \left[ \sum_{i=1}^s m_i(0)w_i - \sum_{j=1}^s E[X_j(t)]w_j - \sum_{k=1}^r \sum_{j=1}^s E[Y_{jk}(t)]w_j \right] \quad (21)$$

$$\begin{aligned} & \text{Var}[X_{s+1}^{\text{gen}}(t)] \\ = & \left[ \sum_{j=1}^s \text{Var}[X_j(t)]w_j^2 \right. \\ & + \sum_{i=1}^s \sum_{j=1}^s \sum_{\substack{d=1 \\ j \neq d}}^s \text{Cov}[X_{ij}(t), X_{id}(t)]w_j w_d \\ & + \sum_{i=1}^s \sum_{j=1}^s \sum_{d=1}^s \sum_{k=1}^r \text{Cov}[X_{ij}(t), Y_{idk}(t)]w_j w_d \\ & + \sum_{i=1}^s \sum_{j=1}^s \sum_{\substack{d=1 \\ j \neq d}}^s \sum_{k=1}^r \sum_{f=1}^r \text{Cov}[Y_{ijk}(t), Y_{idf}(t)]w_j w_d \\ & + \sum_{i=1}^s \sum_{j=1}^s \sum_{\substack{k=1 \\ k \neq f}}^r \sum_{f=1}^r \text{Cov}[Y_{ijk}(t), Y_{ijf}(t)]w_j^2 \\ & \left. + \sum_{k=1}^r \sum_{j=1}^s \text{Var}[Y_{jk}(t)]w_j^2 \right] \quad (22) \end{aligned}$$

The quantities  $E[X_{s+1}^{gen}(t)]$  and  $Var[X_{s+1}^{gen}(t)]$  are obtained as continuous functions of  $t$  and are cumulative; the time derivatives of these quantities yield the mean and variance of the rate of fines generated at any instant  $t$ . Thus, a random amount of fines is generated at each instant  $t$  which either remains within the cell or exits through any of the outlet streams.

A portion of the fines present initially in the cell may remain within at time  $t$  depending on the value of  $p_{s+1,s+1}(0,t)$ . This amount, together with the generated fines that remain in the cell, constitutes the total amount of fines present at time  $t$ , represented by  $X_{s+1}(t)$ . The contribution from the initial population of fines can be obtained from equations (8) and (9). The amount of generated fines remaining in the cell is obtained from the random number of fines generated at each instant and the transition probability. For instance, the expected value of the amount of fines remaining in the cell at time  $t$ , originating from a random quantity of fines generated in the interval  $(\tau, \tau+d\tau)$  between 0 and  $t$ , is

$$\left[ \frac{d}{dt} \{E[X_{s+1}^{gen}(t)]\} \Big|_{t=\tau} d\tau \right] p_{s+1,s+1}(\tau,t)$$

Hence, the expected value of the total amount of generated fines is obtained by integrating the preceding expression between the limits 0 and  $t$ . The expression for the corresponding variance will include both the mean and variance of the random quantity of fines generated,  $X_{s+1}^{gen}(t)$

(see, APPENDIX B). For the total amount of fines in the cell at any time  $t$ ,  $X_{s+1}(t)$ , we therefore, obtain

$$E[X_{s+1}(t)] = m_{s+1}(0)p_{s+1,s+1}(0,t)w_{s+1} + \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{\text{gen}}(\tau)] \right] p_{s+1,s+1}(\tau,t) d\tau \quad (23)$$

$$\begin{aligned} \text{Var}[X_{s+1}(t)] &= m_{s+1}(0)p_{s+1,s+1}(0,t)(1 - p_{s+1,s+1}(0,t))w_{s+1}^2 \\ &+ \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{\text{gen}}(\tau)] \right] p_{s+1,s+1}(\tau,t)(1 - p_{s+1,s+1}(\tau,t)) d\tau \\ &+ \int_0^t \left[ \frac{d}{d\tau} \text{Var}[X_{s+1}^{\text{gen}}(\tau)] \right] (p_{s+1,s+1}(\tau,t))^2 d\tau \end{aligned} \quad (24)$$

For the amount of fines leaving the cell through outlet stream  $k$  by time  $t$ ,  $Y_{s+1,k}(t)$ , we obtain in a similar fashion:

$$E[Y_{s+1,k}(t)] = m_{s+1}(0)q_{s+1,s+1,k}(0,t)w_{s+1} + \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{\text{gen}}(\tau)] \right] q_{s+1,s+1,k}(\tau,t) d\tau \quad (25)$$

$$\begin{aligned} \text{Var}[Y_{s+1,k}(t)] &= m_{s+1}(0)q_{s+1,s+1,k}(0,t)(1 - q_{s+1,s+1,k}(0,t))w_{s+1}^2 \\ &+ \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{\text{gen}}(\tau)] \right] q_{s+1,s+1,k}(\tau,t)(1 - q_{s+1,s+1,k}(\tau,t)) d\tau \\ &+ \int_0^t \left[ \frac{d}{d\tau} \text{Var}[X_{s+1}^{\text{gen}}(\tau)] \right] (q_{s+1,s+1,k}(\tau,t))^2 d\tau \end{aligned} \quad (26)$$



Hence, from a known initial particle size distribution within the cell, the evolution of the particle size distribution can be obtained from these quantities. Similarly, the size distribution of the particles exiting through each outlet stream by time  $t$  can be obtained from the above quantities.

#### Inclusion of Solids Inflow in System Description

The inflow to the cell is considered here as random with a known distribution or moments which may vary among the  $l$  inlets.  $B_{li}^{in}(t)$  is defined as a random variable representing the rate of particles of size  $i$  entering the cell through inlet stream  $l$  at time  $t$ . To analyze the effects of the inflow, the following random variables are defined.

$B_{li}^{in}(0,t)$  = random variable representing the total number of particles of size  $i$  that have entered the cell through inlet stream  $l$  by time  $t$ .

$B_{lijk}(t)$  = random variable representing the total number of particles of size  $i$  entering the cell through inlet stream  $l$  at time  $t=0$ , that have resulted in particles of size  $j$  and exited through outlet stream  $k$  by time  $t$ ,

$B_{lij}(t)$  = random variable representing the number of particles of size  $i$  entering the cell through inlet stream  $l$  at time  $t=0$ , that have resulted in particles of size  $j$  within the cell at time  $t$ ,

$B_{lj}(t)$  = random variable representing the total number of particles entering through inlet stream  $l$  at time  $t=0$ , that

have resulted in particles of size  $j$  within the cell at time  $t$ .

$B_{ijk}(t)$  = random variable representing the total number of particles entering the cell through inlet stream  $i$  at time  $t=0$ , that have resulted in particles of size  $j$  and exited through outlet stream  $k$  by time  $t$ .

It can be seen that

$$B_{lj}(t) = \sum_{i=1}^j B_{lij}(t) \quad (27)$$

$$B_{ijk}(t) = \sum_{i=1}^j B_{lijk}(t) \quad (28)$$

Any particle entering the cell can undergo a reduction in size, exit from the cell or remain within it during time  $t$ . Assuming these events to occur independently, the various statistical quantities for the inflow are derived in a fashion similar to the derivation of equations (23) through (26) (see APPENDIX B).

$$E[B_{lij}(t)] = \int_0^t E[B_{li}^{in}(\tau)] p_{ij}(\tau, t) d\tau \quad (29)$$

$$\begin{aligned} \text{Var}[B_{lij}(t)] = & \int_0^t E[B_{li}^{in}(\tau)] p_{ij}(\tau, t) (1 - p_{ij}(\tau, t)) d\tau \\ & + \int_0^t \text{Var}[B_{li}^{in}(\tau)] (p_{ij}(\tau, t))^2 d\tau \quad (30) \end{aligned}$$

$$\text{Cov}[B_{li}^{in}(0, t), B_{lij}(t)] = \int_0^t \text{Var}[B_{li}^{in}(\tau)] p_{ij}(\tau, t) d\tau \quad (31)$$

$$\begin{aligned} \text{Cov}[B_{lij}(t), B_{lid}(t)] = & - \int_0^t E[B_{li}^{in}(\tau)] p_{ij}(\tau, t) p_{id}(\tau, t) d\tau \\ & + \int_0^t \text{Var}[B_{li}^{in}(\tau)] p_{ij}(\tau, t) p_{id}(\tau, t) d\tau \quad (32) \end{aligned}$$

$$E[B_{lijk}(t)] = \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) d\tau \quad (33)$$

$$\begin{aligned} \text{Var}[B_{lijk}(t)] = & \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) (1 - q_{ijk}(\tau, t)) d\tau \\ & + \int_0^t \text{Var}[B_{li}^{in}(\tau)] (q_{ijk}(\tau, t))^2 d\tau \quad (34) \end{aligned}$$

$$\text{Cov}[B_{li}^{in}(0, t), B_{lijk}(t)] = \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) d\tau \quad (35)$$

$$\begin{aligned} & \text{Cov}[B_{lijk}(t), B_{lidf}(t)] \\ = & - \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{idf}(\tau, t) d\tau \\ & + \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{idf}(\tau, t) d\tau \quad (36) \end{aligned}$$

$$\begin{aligned} & \text{Cov}[B_{lijk}(t), B_{lijf}(t)] \\ = & - \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{ijf}(\tau, t) d\tau \\ & + \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{ijf}(\tau, t) d\tau, \quad k \neq f \end{aligned}$$

(37)

$$\text{Cov}[B_{lij}(t), B_{lidk}(t)]$$

$$= - \int_0^t E[B_{li}^{in}(\tau)] p_{ij}(\tau, t) q_{idk}(\tau, t) d\tau + \int_0^t \text{Var}[B_{li}^{in}(\tau)] p_{ij}(\tau, t) q_{idk}(\tau, t) d\tau, \quad (38)$$

$$i=1, 2, \dots, s; j, d=i, i+1, \dots, s+1; j \neq d; l=1, 2, \dots, b; k=1, 2, \dots, r$$

In the case of fines, the results similar to equations (21) through (26) are obtained as:

$$E[B_{l, s+1}^{gen}(t)] = \sum_{i=1}^s w_i \int_0^t E[B_{li}^{in}(\tau)] d\tau - \sum_{j=1}^s w_j E[B_{lj}(t)] - \sum_{j=1}^s \sum_{k=1}^r w_j E[B_{ljk}(t)] \quad (39)$$

$$\begin{aligned} & \text{Var}[B_{l, s+1}^{gen}(t)] \\ = & \sum_{i=1}^s w_i^2 \int_0^t \text{Var}[B_{li}^{in}(\tau)] d\tau + \sum_{j=1}^s \text{Var}[B_{lj}(t)] w_j^2 \\ & + \sum_{i=1}^s \sum_{j=i}^s \text{Cov}[B_{li}^{in}(0, t), B_{lij}(t)] w_i w_j \\ & + \sum_{i=1}^s \sum_{j=i}^s \sum_{d=i}^s \text{Cov}[B_{lij}(t), B_{lid}(t)] w_j w_d \\ & + \sum_{i=1}^s \sum_{j=i}^s \sum_{k=1}^r \text{Cov}[B_{li}^{in}(t), B_{lijfk}(t)] w_i w_j \\ & + \sum_{i=1}^s \sum_{j=i}^s \sum_{k=1}^r \sum_{f=k}^r \text{Cov}[B_{li}^{in}(t), B_{lijfk}(t)] w_j^2 \end{aligned}$$

$$\begin{aligned}
& + \sum_{i=1}^s \sum_{j=1}^s \sum_{d=i}^s \sum_{k=1}^r \text{Cov}[B_{ijk}(t), B_{idk}(t)] w_j w_d \\
& \quad j \neq d \\
& + \sum_{i=1}^s \sum_{j=1}^s \sum_{d=i}^s \sum_{k=1}^r \text{Cov}[B_{ij}(t), B_{idk}(t)] w_j w_d \\
& \quad j \neq d \\
& + \sum_{j=1}^s \sum_{k=1}^r \text{Var}[B_{ijk}(t)] w_j^2
\end{aligned} \tag{40}$$

Hence,

$$\begin{aligned}
E[B_{l,s+1}(t)] &= w_{s+1} \int_0^t E[B_{l,s+1}^{\text{in}}(\tau)] p_{s+1,s+1}(\tau, t) d\tau \\
&+ \int_0^t \left[ \frac{d}{d\tau} E[B_{l,s+1}^{\text{gen}}(\tau)] \right] (p_{s+1,s+1}(\tau, t)) d\tau \quad (41)
\end{aligned}$$

$$\begin{aligned}
& \text{Var}[B_{l,s+1}(t)] \\
&= w_{s+1}^2 \int_0^t E[B_{l,s+1}^{\text{in}}(\tau)] p_{s+1,s+1}(\tau, t) (1 - p_{s+1,s+1}(\tau, t)) d\tau \\
&+ w_{s+1}^2 \int_0^t \text{Var}[B_{l,s+1}^{\text{in}}(\tau)] (p_{s+1,s+1}(\tau, t))^2 d\tau \\
&+ \int_0^t \left[ \frac{d}{d\tau} E[B_{l,s+1}^{\text{gen}}(\tau)] \right] p_{s+1,s+1}(\tau, t) (1 - p_{s+1,s+1}(\tau, t)) d\tau \\
&+ \int_0^t \left[ \frac{d}{d\tau} \text{Var}[B_{l,s+1}^{\text{gen}}(\tau)] \right] (p_{s+1,s+1}(\tau, t))^2 d\tau \quad (42)
\end{aligned}$$

where  $B_{l,s+1}^{\text{in}}(t)$  represents the rate of inflow of fines through inlet stream  $l$ . Similarly, inclusion of the inflow of fines yields

$$E[B_{l,s+1,k}(t)] = \int_0^t E[B_{l,s+1}^{\text{in}}(\tau)] q_{s+1,s+1,k}(\tau, t) d\tau$$

$$+ \int_0^t \left[ \frac{d}{d\tau} E[B_{l,s+1}^{\text{gen}}(\tau)] \right] q_{s+1,s+1,k}(\tau, t) d\tau \quad (43)$$

$$\begin{aligned} & \text{Var}[B_{l,s+1,k}(t)] \\ = & w_{s+1} \int_0^t E[B_{l,s+1}^{\text{in}}(\tau)] q_{s+1,s+1,k}(\tau, t) (1 - q_{s+1,s+1,k}(\tau, t)) d\tau \\ & + w_{s+1}^2 \int_0^t \text{Var}[B_{l,s+1}^{\text{in}}(\tau)] (q_{s+1,s+1,k}(\tau, t))^2 d\tau \\ & + \int_0^t \left[ \frac{d}{d\tau} E[B_{l,s+1}^{\text{gen}}(\tau)] \right] (q_{s+1,s+1,k}(\tau, t) (1 - q_{s+1,s+1,k}(\tau, t))) d\tau \\ & + \int_0^t \left[ \frac{d}{d\tau} \text{Var}[B_{l,s+1}^{\text{gen}}(\tau)] \right] (q_{s+1,s+1,k}(\tau, t))^2 d\tau \quad (44) \end{aligned}$$

Thus, for the system with inflow, attrition in the cell and outflow, the equations for the total number of particles of various sizes and the amount of fines are obtained as:

$$E[X_j^{\text{tot}}(t)] = E[X_j(t)] + \sum_{l=1}^b E[B_{lj}(t)] \quad (45)$$

$$\text{Var}[X_j^{\text{tot}}(t)] = \text{Var}[X_j(t)] + \sum_{l=1}^b \text{Var}[B_{lj}(t)] \quad (46)$$

$$E[Y_{jk}^{\text{tot}}(t)] = E[Y_{jk}(t)] + \sum_{l=1}^b E[B_{ljk}(t)] \quad (47)$$

$$\text{Var}[Y_{jk}^{\text{tot}}(t)] = \text{Var}[Y_{jk}(t)] + \sum_{l=1}^b \text{Var}[B_{ljk}(t)], \quad (48)$$

$$j = 1, 2, \dots, s+1; k = 1, 2, \dots, r$$

The superscript "tot" refers to the entire system.

## APPLICATIONS

The mechanics of size reduction by abrasion or attrition are observed frequently in many particulate processes. In the following, applications of the general model to two specific areas are outlined.

### Chemical Attrition in Fluidization

Attrition in fluidized beds can be classified into two modes, mechanical attrition, arising from the physical impact of collisions between particles or with the bed walls, and chemical attrition, occurring due to combustion or reaction that tends to corrode the surface of the particulate material (see, e.g., Chirone et al., 1985). It has been observed (see, e.g., Doheim et al., 1976) that depending on the properties of the material undergoing combustion, chemical attrition may be even an order of magnitude higher than purely mechanical attrition, i.e., chemical attrition predominates when a reaction occurs in the bed. The present general cell model is applicable to such systems where chemical attrition predominates. An alternative treatment is required when studying mechanical attrition dealing with interparticle and particle-wall collisions.

By considering that the particles undergo abrasion as chemical reaction proceeds, akin to the shrinking particle model, the transition intensities pertaining to the size degradation assume the general form (see, e.g., Lin et al., 1980, and Merrick and Highley, 1974)

$$v_{i,i+1} = \beta_i (U_f - U_{mf}) \quad (49)$$

where  $U_f$  is the superficial gas velocity,  $U_{mf}$ , the minimum fluidization velocity and  $\beta_1$ , a parameter which is specific to the particle size and includes the material and bed properties. The exit transition intensities  $\mu_{jk}$  depend on the flow pattern. Considering the bed as a continuous stirred tank reactor (CSTR), it may be assumed that the particles have equal probabilities of leaving the bed. The exit transition intensities,  $\mu_{jk}$ , may then be evaluated as  $w/W$ , where  $w$  is the outflow and  $W$ , the total particle bed weight. Entrainment streams rarely contain large sized particles and any empirical correlation, yielding the entrainment rate as a function of particle size, can be utilized in determining the form of the exit transition intensity.

The distribution of particle inflow rates for each size is required. If particles are fed from a hopper, this distribution may be regarded as Poisson. The distribution of the inflow rate from a recycle stream or from another bed can be evaluated by applying the general model to that particular bed. The rate of particles of each size entering the bed through a feed stream (from a continuous classifier, for instance) may be described by an appropriate normal distribution.

#### Particle Size Reduction by Abrasion

Three distinct breakage mechanisms can be considered to prevail during the size reduction operations in mineral processing. Particles in a ball mill, for instance, may undergo abrasion, i.e., the surface of the particle is constantly abraded generating fines; particles may undergo chipping, especially if irregular in shape or the particles may



break into two or more fragments (see, e.g., Austin and Barahona, 1986). The size reduction processes are known to display a stochastic nature and numerous attempts have been made to study the random disintegration of particles during the size reduction operation (see, e.g., Rose, 1957, and Auer, 1978). The present model can be applied to systems where the mode of particle disintegration is predominantly abrasion.

By assuming that each particle has a certain probability of undergoing abrasion in a small time interval, independent of the other particles, the transition intensities for the change in state (size) of particles from one size to the next smaller size can be obtained in a general form as

$$v_{i,i+1} = \kappa_i S_i \quad (50)$$

where  $S_i$  is the rate of selection for abrasion and  $\kappa_i$  is a constant dependent on material and mill properties (see, e.g., Klimpel and Austin, 1984). It has been observed that larger particles tend to abrade more than smaller ones; this can be stated as  $v_{12} > v_{23} \dots$ . In considering a continuous flow system, the residence time of the solids needs to be included. Different flow models have been applied to describe the effect of material flow on the product size distribution (see, e.g., Austin et al., 1983). The finite stage transport model has been found adequate in describing the flow of solids in a mill (see, e.g., Rogers and Gardner, 1979). The exit transition intensities,  $\mu_{jk}$ , can be derived easily based on the model utilized to describe the solids flow.

# NUMERICAL SIMULATION

Multistage fluidized bed operations involving reactions are commonly encountered in the chemical process industry (see, e.g., Kunii and Levenspiel, 1969). The application of the model is illustrated with an example of a two-stage fluidized-bed combustion operation as shown in Figure 3. Particles of two size ranges denoted by 1 and 2 with the mean weights of 500 and 250 mg, respectively, are present in the two beds initially in the absence of fines, denoted by size range 3. The beds are allowed to attain the incipient fluidization state until which time no attrition is assumed to occur. Solids of the two sizes enter the first bed at a Poisson rate from a hopper. The outflow of the first bed serves as the feed to the second. Complete mixing of the solids in each bed and independence of particle size are assumed in deriving values for the exit transition intensities. Entrainment of the fines is assumed to occur in both beds. The transition intensities for the first bed assumes the following values:

$$v_{ij} = \begin{matrix} & \begin{matrix} j \\ 1 \quad 2 \quad 3 \end{matrix} \\ \begin{matrix} i \\ 1 \\ 2 \\ 3 \end{matrix} & \begin{bmatrix} -0.07 & 0.05 & 0 \\ 0 & -0.05 & 0.03 \\ 0 & 0 & -0.06 \end{bmatrix} \end{matrix}$$

$$\mu_{jk} = \begin{matrix} & \begin{matrix} k \\ 1 \quad 2 \end{matrix} \\ \begin{matrix} j \\ 1 \\ 2 \\ 3 \end{matrix} & \begin{bmatrix} 0.02 & 0 \\ 0.02 & 0 \\ 0.02 & 0.04 \end{bmatrix} \end{matrix}$$

The second bed is assumed to be operated under such conditions that the transition intensities are the same as bed 1 for size 1 and size 2 particles while all fines are entrained immediately. Thus, the transition intensities for the second bed assume the following values:

$$v_{ij} = \begin{matrix} & \begin{matrix} j \\ 1 \quad 2 \quad 3 \end{matrix} \\ \begin{matrix} i \\ 1 \\ 2 \\ 3 \end{matrix} & \begin{bmatrix} -0.07 & 0.05 & 0 \\ 0 & -0.05 & 0.03 \\ 0 & 0 & -1.0 \end{bmatrix} \end{matrix}$$

$$\mu_{jk} = \begin{matrix} & \begin{matrix} j \\ 1 \quad 2 \end{matrix} \\ \begin{matrix} i \\ 1 \\ 2 \\ 3 \end{matrix} & \begin{bmatrix} 0.02 & 0 \\ 0.02 & 0 \\ 0 & 1.0 \end{bmatrix} \end{matrix}$$

The initial population sizes and inflow rates are given by

$$n_1^1(0) = 1000; \quad n_2^1(0) = 1000; \quad n_3^1(0) = 0;$$

$$n_1^2(0) = 2000; \quad n_2^2(0) = 2000; \quad n_3^2(0) = 0;$$

$$E[B_{11}^{lin}(\tau)] = 100 \text{ time}^{-1}; \quad \text{Var}[B_{11}^{lin}(\tau)] = 100 \text{ time}^{-1}.$$

where the superscript indicates bed number. The transition probabilities,  $p_{ij}(0,t)$  and  $q_{ijk}(0,t)$ , are derived from the transition intensities as detailed previously. These are listed in Tables 1 and 2.

The feed to the second bed is calculated from the evolution of the particle size distribution in the first bed. The fines generated from attrition are obtained in terms of mass; the corresponding number can be obtained from the ratio of the mass to the mean weight. The number of particles exiting a bed is cumulative; it is the number of particles leaving the bed by time  $t$ . These values and the temporal changes in the particle populations of the different sizes in the beds are plotted in Figures 4 through 27. The values of the various quantities involving fines, i.e.,  $E[X_3(t)]$ ,  $\text{Var}[X_3(t)]$ ,  $E[Y_{31}(t)]$ ,  $E[B_{13}(t)]$ , etc. are

plotted in units of mass. The amount of fines in the second bed resulting from inflow and attrition is presented in Figures 22 and 27 as the cumulative amount entrained by time  $t$ .

## CONCLUSIONS

The general cell model describes the stochastic nature of attrition, providing estimates of the fluctuations arising thereof. The modularity of the model enhances its application to continuous and multiple flow systems. The numerical example indicates that larger fluctuations can be expected in the population of the smaller particles into which the larger or coarser particles abrade. Fluctuations in the inflow are magnified to a considerable extent; it may be as high as 10 to 15 percent by weight in the case of fines. This has a great impact on elutriation studies.

The probabilistic considerations involved in deriving the model also indicate that the fluctuations in the populations of various sizes decrease as the intensity of attrition for a given size tends to unity; this corresponds to the deterministic limit. The probabilities of transition of particles between states have been derived from binomial consideration; this also indicates that the probability distribution of the size of the particle population approaches the Poisson distribution asymptotically as the distribution evolves. That is, the mean and the variance of the number of particles coincide, implying that the standard deviation is of the order of the square root of the particle population size. This may or may not be significant depending on the system under study. For instance, the fluctuations in the amount of fines produced within the bed in fluidization may not be significant with respect to

the bed size. This may not greatly affect the bed characteristics but is important when controlling the emission of fines from the bed under environmental consideration.

## NOTATION

B	random variable representing the rate of particles entering the cell
b	number of inlet streams
d	subscript referring to size ranges within the cell
f	subscript referring to outlet streams
i	subscript referring to size ranges within the cell
j	subscript referring to size ranges within the cell or in outlet streams
k	subscript referring to outlet streams
l	subscript referring to inlet streams
p	probability referring to transitions between size ranges within the cell
q	probability referring to transitions between size ranges within the cell
r	number of outlet streams
S	intensity of selection for abrasion, time <sup>-1</sup>
s	number of size ranges excluding fines
s+1	the size range of fines
t	time
w	mean weight of a size range
X	random variable referring to particle population size within the cell
Y	random variable referring to particle population size in the outlet streams

## Greek Letters

$\beta$	size specific constant in the transition intensity of attrition in fluidized beds, length <sup>-1</sup>
$\kappa$	size specific constant in the transition intensity of abrasion

$\mu$  transition intensity of exit from the cell,  $\text{time}^{-1}$   
 $\tau$  time  
 $\nu$  intensity of transition between size ranges within the cell,  
 $\text{time}^{-1}$

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# APPENDIX A. DERIVATION OF TRANSITION PROBABILITIES

Consider a time interval  $(\tau, t)$  and a fixed time  $\xi$  between  $\tau$  and  $t$ . The Markovian assumption implies that the future transitions of the particle are independent of the past transitions. It follows, then, that the probability that a particle of size  $l$  will be of size  $c$  at time  $\xi$  and is of size  $j$  at time  $t$  is

$$p_{lc}(\tau, \xi) p_{cj}(\xi, t), \quad i=1, 2, \dots, s;$$

$$c, j=i+1, \dots, s+1$$

Since the events corresponding to that described above are mutually exclusive, for different  $c$ , we have

$$p_{ij}(\tau, t) = \sum_{c=i}^{s+1} p_{lc}(\tau, \xi) p_{cj}(\xi, t) \quad (A1)$$

for all  $i=1, 2, \dots, s$  and  $j=i+1, \dots, s+1$

The above equation is a form of the Chapman-Kolmogorov equation describing a Markov process. The differential equations for  $p_{ij}(\tau, t)$  are obtained by considering two contiguous time intervals  $(\tau, t)$  and  $(t, t+\Delta t)$ , and  $p_{ij}(\tau, t+\Delta t)$ . Using the definitions of the transition intensities, we obtain:

$$p_{i1}(\tau, t+\Delta t) = p_{i1}(\tau, t)(1+u_{i1}\Delta t) + o(\Delta t) \quad (A2)$$

$$p_{ij}(\tau, t+\Delta t) = p_{i1}(\tau, t)u_{ij}\Delta t$$

$$+ p_{ij}(\tau, t)(1+u_{jj}\Delta t) + o(\Delta t) \quad (A3)$$

where any other kind of transitions (more than one transition during the interval  $(t, t+\Delta t)$  occurs with a probability of the order of  $o(\Delta t)$ . Rearranging the above equations to form difference quotients and taking the limit  $\Delta t \rightarrow 0$ , we obtain a system of linear first order differential equation for the probability balance,

$$\frac{\partial}{\partial t} p_{ij}(\tau, t) = \sum_{c=1}^{s+1} p_{ic}(\tau, t) u_{cj} \quad (\text{A-4})$$

for all  $i=1, 2, \dots, s+1$ ;  $j=c, c+1, \dots, s+1$

The transition intensities can be represented in matrix form as

$$V = \begin{bmatrix} u_{11} & u_{12} & 0 & 0 & \dots & 0 & 0 \\ 0 & u_{22} & u_{23} & 0 & \dots & 0 & 0 \\ 0 & 0 & u_{33} & u_{34} & \dots & 0 & 0 \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 0 & 0 & 0 & 0 & \dots & u_{s,s} & u_{s,s+1} \\ 0 & 0 & 0 & 0 & \dots & 0 & u_{s+1,s+1} \end{bmatrix}$$

The solution can be obtained in terms of the eigenvalues  $\rho_1, \rho_2, \dots, \rho_s, \rho_{s+1}$ ,

which are the roots of the characteristic equation  $|\rho I - V| = 0$ , assuming a

non-trivial solution to exist for the given problem.

The solution for the case of distinct eigenvalues is (see, e.g., Chiang, 1980):

$$p_{ij}(\tau, t) = \sum_{m=1}^{s+1} \frac{A'_{ij}(\rho_m) e^{\rho_m(t-\tau)}}{\prod_{\substack{n=1 \\ n \neq m}}^{s+1} (\rho_m - \rho_n)} \quad (\text{A5a})$$

or

$$p_{ij}(\tau, t) = \sum_{m=1}^{s+1} \frac{A_i(\rho_m) T_{jm}(p) e^{\rho_m(t-\tau)}}{T(p)} \quad (\text{A5b})$$

where  $V'$  is the transpose of  $V$ ,  $A'_{ij}(\rho_m)$  is the  $(j,i)$ th cofactor of the characteristic matrix of  $V$ , for  $\rho = \rho_m$ ,  $A(\rho_m) = (\rho_m I - V)$ , and  $T_{jm}(p)$  is the cofactor of the non-singular matrix

$$T(p) = \begin{bmatrix} A_{p1}(\rho_1) & \dots & A_{p1}(\rho_{s+1}) \\ \vdots & & \vdots \\ A_{ps+1}(\rho_1) & \dots & A_{ps+1}(\rho_{s+1}) \end{bmatrix}$$

The second equation can be compactly represented in matrix notation as

$$P(\tau, t) = T(p)E(t-\tau)T^{-1}(p) \quad (A6)$$

where  $E(t-\tau)$  is a diagonal matrix

$$E(t-\tau) = \begin{bmatrix} e^{\rho_1(t-\tau)} & 0 & \dots & 0 \\ 0 & e^{\rho_2(t-\tau)} & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \dots & e^{\rho_{s+1}(t-\tau)} \end{bmatrix}$$

Solutions exist for the case of multiple eigenvalues, which are discussed elsewhere (see, e.g., Chiang, 1980).

APPENDIX B. DERIVATION OF RELATIONS TO HANDLE RANDOM INITIAL DISTRIBUTION  
AND RANDOM INFLOW OF PARTICLES

Considering  $m_i(0)$  particles of size  $i$  initially in the cell leads to the expressions for the mean and variance of the various  $\{X_{ij}(t)\}$  and  $\{Y_{ijk}(t)\}$  [see equations (8) through (11) in the text]. Let us denote the random number of particles of size  $i$  present in the cell initially by  $M_i(0)$ . Moreover, the probability distribution of  $M_i(0)$  or its moments are assumed to be known. Then, the expressions for the expected value and variance for  $\{X_{ij}(t)\}$  or  $\{Y_{ijk}(t)\}$  are obtained as follows:

Consider a random number of particles of a single size present initially with known mean and variance,  $E[M_i(0)]$  and  $\text{Var}[M_i(0)]$ , respectively. These particles distribute themselves among the  $i, i+1, \dots, s+1$ ;  $k=1, 2, \dots, r$  states at any time  $t$ . Let  $X$  and  $Y$  be random variables. Furthermore let  $E[X|Y]$  be a function of the random variable  $X$  whose value at  $Y=y$  is  $E[X|Y=y]$ . Note that  $E[X|Y]$  is itself a random variable and from the property of conditional expectation

$$E[X] = E[E[X|Y]] \quad (B1)$$

Applying the above relationship to  $X_{ij}(t)$ ,  $Y_{ijk}(t)$ , and  $M_i(0)$

$$E[X_{ij}(t)] = E[E[X_{ij}(t)|M_i(0)]] \quad (B2)$$

$$E[Y_{ijk}(t)] = E[E[Y_{ijk}(t)|M_i(0)]] \quad (B3)$$

The derivation remains essentially the same with respect to  $\{X_{ij}(t)\}$  or  $\{Y_{ijk}(t)\}$ . Hence, in the following, only  $\{X_{ij}(t)\}$  is considered but the final expressions for both  $\{X_{ij}(t)\}$  and  $\{Y_{ijk}(t)\}$  are provided.

$$E[X_{ij}(t)|M_i(0)] = M_i(0)p_{ij}(0,t) \quad (B4)$$

$$E[E[X_{ij}(t)|M_i(0)]] = E[M_i(0)]p_{ij}(0,t) \quad (B5)$$

Thus,

$$E[X_{ij}(t)] = E[M_i(0)]p_{ij}(0,t) \quad (B6)$$

By definition, the expression for the variance is obtained as:

$$\text{Var}[X_{ij}(t)] = E[(X_{ij}(t))^2] - (E[X_{ij}(t)])^2 \quad (B7)$$

where each of the R.H.S. terms is conditional on  $M_i(0)$ . Using equation (B1)

$$E[(X_{ij}(t))^2] = E[E[(X_{ij}(t))^2|M_i(0)]] \quad (B8)$$

From the definition of moments of a random variable,  $\text{Var}[Y] = E[Y^2] - (E[Y])^2$ , where  $Y$  is any random variable. Applying this relation to the R.H.S. term in equation (B8) yields

$$\begin{aligned} E[(X_{ij}(t))^2] &= E[M_i(0)]p_{ij}(0,t)(1-p_{ij}(0,t)) \\ &\quad + E[(M_i(0))^2](p_{ij}(0,t))^2 \end{aligned} \quad (B9)$$

On substitution into equation (B7) and further reduction, the expression for  $\text{Var}[X_{ij}(t)]$  is obtained as

$$\begin{aligned} \text{Var}[X_{ij}(t)] &= E[M_i(0)]p_{ij}(0,t)(1-p_{ij}(0,t)) \\ &\quad + \text{Var}[M_i(0)](p_{ij}(0,t))^2 \end{aligned} \quad (B10)$$

The covariance of two random variables  $X$  and  $Y$  can be expressed as

$$\text{Cov}[X,Y] = E[XY] - E[X]E[Y] \quad (B11)$$

The covariance of  $X$  and  $Y$  is a measure of a linear relationship of  $X$  and  $Y$  in the sense that it will be positive when  $(X-E[X])$  and  $(Y-E[Y])$  assume the

same sign with high probability. It will be negative when  $(X-E[X])$  and  $(Y-E[Y])$  assume opposite signs with high probability. In the physical sense, the covariance of  $M_i(0)$  and  $X_{ij}(t)$  is positive as an increase in  $M_i(0)$  results in a corresponding increase in  $X_{ij}(t)$ . The covariance of  $X_{ij}(t)$  and  $X_{id}(t)$ , however, is negative as a positive deviation in  $X_{ij}(t)$  results in a negative deviation in  $X_{id}(t)$ . This arises from the condition

$$M_i(0) = \sum_{j=1}^{s+1} X_{ij}(t)$$

The covariance of  $M_i(0)$  and  $X_{ij}(t)$  is derived in the following manner.

$$\text{Cov}[M_i(0), X_{ij}(t)] = E[M_i(0)X_{ij}(t)] - E[M_i(0)]E[X_{ij}(t)] \quad (\text{B12})$$

The two terms on the R.H.S. are conditional on  $M_i(0)$ . The first term is expressed as

$$\begin{aligned} E[M_i(0)X_{ij}(t)] &= E[E[M_i(0)X_{ij}(t)] | M_i(0)] \\ &= E[M_i(0)E[X_{ij}(t)] | M_i(0)] \\ &= E[M_i(0)^2 p_{ij}(0, t)] \\ &= E[M_i(0)^2] p_{ij}(0, t) \end{aligned} \quad (\text{B13})$$

Substituting the expression given by equation (B4) for  $E[X_{ij}(t)]$  on the R.H.S. yields

$$\begin{aligned} \text{Cov}[M_i(0), X_{ij}(t)] &= E[M_i(0)^2] p_{ij}(0, t) - E[M_i(0)]^2 p_{ij}(0, t) \\ &= \text{Var}[M_i(0)] p_{ij}(0, t) \end{aligned} \quad (\text{B14})$$

In the case of the random variables  $X_{ij}(t)$  and  $X_{id}(t)$ , the covariance assumes the form

$$\text{Cov}[X_{ij}(t), X_{id}(t)] = E[X_{ij}(t)X_{id}(t)] - E[X_{ij}(t)]E[X_{id}(t)] \quad (B15)$$

The first term on the R.H.S. conditional on a random initial distribution  $M_i(0)$  is expressed as

$$E[X_{ij}(t)X_{id}(t)] = E[E[X_{ij}(t)X_{id}(t)|M_i(0)]] \quad (B16)$$

Equations (B11) and (B6) are utilized to express the R.H.S. as

$$\begin{aligned} & E[E[X_{ij}(t)X_{id}(t)|M_i(0)]] \\ &= E[\{\text{Cov}[X_{ij}(t)X_{id}(t)] + E[X_{ij}(t)] E[X_{id}(t)]\}|M_i(0)] \\ &= -E[M_i(0)]p_{ij}(0,t)p_{id}(0,t) + E[M_i(0)^2]p_{ij}(0,t)p_{id}(0,t) \end{aligned} \quad (B17)$$

On substituting equation (B17) into equation (B16) and simplifying equation (B15) we obtain an expression for the covariance of  $X_{ij}(t)$  and  $X_{id}(t)$  as

$$\begin{aligned} \text{Cov}[X_{ij}(t), X_{id}(t)] &= -E[M_i(0)]p_{ij}(0,t)p_{id}(0,t) \\ &\quad + \text{Var}[M_i(0)]p_{ij}(0,t)p_{id}(0,t) \end{aligned} \quad (B18)$$

Equations (B6), (B10), and (B18) are the required expressions with regard to  $\{X_{ij}(t)\}$ . The corresponding equations with respect to  $\{Y_{ijk}(t)\}$  are obtained in a similar fashion as

$$E[Y_{ijk}(t)] = E[M_i(0)]q_{ijk}(0,t) \quad (B19)$$

$$\begin{aligned} \text{Var}[Y_{ijk}(t)] &= E[M_i(0)]q_{ijk}(0,t)(1-q_{ijk}(0,t)) \\ &\quad + \text{Var}[M_i(0)](q_{ijk}(0,t))^2 \end{aligned} \quad (B20)$$

Similar equations result for the covariances of  $Y_{ijk}(t)$  and  $Y_{idk}(t)$ , and  $X_{ij}(t)$  and  $Y_{idk}(t)$ , which can be written as

$$\text{Cov}[Y_{ijk}(t), Y_{idk}(t)] = -E[M_i(0)]q_{ijk}(0,t)q_{idk}(0,t)$$



$$+ \text{Var}[M_i(0)]q_{ijk}(0,t)q_{idk}(0,t) \quad (\text{B21})$$

$$\begin{aligned} \text{Cov}[Y_{ijf}(t), Y_{ijf}(t)] &= -E[M_i(0)]q_{ijk}(0,t)q_{ijf}(0,t) \\ &+ \text{Var}[M_i(0)]q_{ijk}(0,t)q_{ijf}(0,t) \end{aligned} \quad (\text{B22})$$

$$\begin{aligned} \text{Cov}[X_{ij}(t), Y_{idk}(t)] &= -E[M_i(0)]p_{ij}(0,t)q_{idk}(0,t) \\ &+ \text{Var}[M_i(0)]p_{ij}(0,t)q_{idk}(0,t) \end{aligned} \quad (\text{B23})$$

It must be noted that the covariances exist only for  $j=d$ ; they are otherwise termed as variances.

The above derivation can be extended to the case of an inflow of particles whose distribution may change at each instant  $t$ . The groups of particles arriving at each instant distribute themselves among the various sizes and exit streams; hence, for an inflow denoted by  $B_{li}^{\text{in}}(\tau)$ , a random variable representing the number of particles of size  $i$  arriving at the cell at time  $\tau$ , with  $E[B_{li}^{\text{in}}(\tau)]$  and  $\text{Var}[B_{li}^{\text{in}}(\tau)]$  known from the inlet stream  $l$  as defined in the text, the required expressions are obtained as:

$$E[B_{lij}(t)] = \int_0^t E[B_{li}^{\text{in}}(\tau)]p_{ij}(\tau,t)d\tau \quad (\text{B24})$$

$$\begin{aligned} \text{Var}[B_{lij}(t)] &= \int_0^t E[B_{li}^{\text{in}}(\tau)]p_{ij}(\tau,t)(1-p_{ij}(\tau,t))d\tau \\ &+ \int_0^t \text{Var}[B_{li}^{\text{in}}(\tau)](p_{ij}(\tau,t))^2d\tau \end{aligned} \quad (\text{B25})$$

$$\text{Cov}[B_{li}^{\text{in}}(0,t), B_{lij}(t)] = \int_0^t \text{Var}[B_{li}^{\text{in}}(\tau)]p_{ij}(\tau,t)d\tau \quad (\text{B26})$$

$$\begin{aligned} &\text{Cov}[B_{lij}(t), B_{lid}(t)] \\ &= - \int_0^t E[B_{li}^{\text{in}}(\tau)]p_{ij}(\tau,t)p_{id}(\tau,t)d\tau \end{aligned}$$

$$+ \int_0^t \text{Var}[B_{li}^{in}(\tau)] p_{ij}(\tau, t) p_{id}(\tau, t) d\tau \quad (B27)$$

$$E[B_{lijk}(t)] = \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) d\tau \quad (B28)$$

$$\begin{aligned} \text{Var}[B_{lijk}(t)] &= \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) (1 - q_{ijk}(\tau, t)) d\tau \\ &\quad + \int_0^t \text{Var}[B_{li}^{in}(\tau)] (q_{ijk}(\tau, t))^2 d\tau \end{aligned} \quad (B29)$$

$$\text{Cov}[B_{li}^{in}(0, t), B_{lijk}(t)] = \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) d\tau \quad (B30)$$

$$\begin{aligned} &\text{Cov}[B_{lijk}(t), B_{lidf}(t)] \\ &= - \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{idf}(\tau, t) d\tau \\ &\quad + \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{idf}(\tau, t) d\tau, \quad j \neq d \end{aligned} \quad (B31)$$

$$\begin{aligned} &\text{Cov}[B_{lijk}(t), B_{lijf}(t)] \\ &= - \int_0^t E[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{ijf}(\tau, t) d\tau \\ &\quad + \int_0^t \text{Var}[B_{li}^{in}(\tau)] q_{ijk}(\tau, t) q_{ijf}(\tau, t) d\tau, \quad k \neq f \end{aligned} \quad (B32)$$

$$\begin{aligned} &\text{Cov}[B_{lij}(t), B_{lidk}(t)] \\ &= - \int_0^t E[B_{li}^{in}(\tau)] p_{ij}(\tau, t) q_{idk}(\tau, t) d\tau \\ &\quad + \int_0^t \text{Var}[B_{li}^{in}(\tau)] p_{ij}(\tau, t) q_{idk}(\tau, t) d\tau \end{aligned} \quad (B33)$$

$B_{li}^{in}(0, t)$ , occurring in equations (B26) and (B30), is the random variable representing the total inflow of particles of size  $i$  from inlet stream  $l$  by time  $t$ .

The above derivation can be applied to obtain the total amount of fines generated by substituting  $\frac{d}{dt}E[X_{s+1}^{gen}(t)]$  for  $E[B_{li}^{in}(\tau)]$  and  $\frac{d}{dt}\text{Var}[X_{s+1}^{gen}(t)]$  for  $\text{Var}[B_{li}^{in}(\tau)]$ , in the corresponding equations. Together with the expressions for an initial number of fines present, the results are:

$$E[X_{s+1}(t)] = m_{s+1}(0)p_{s+1,s+1}(0, t) + \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{gen}(\tau)] \right] p_{s+1,s+1}(\tau, t) d\tau \quad (B34)$$

$$\begin{aligned} \text{Var}[X_{s+1}(t)] &= m_{s+1}(0)p_{s+1,s+1}(0, t)(1 - p_{s+1,s+1}(0, t)) \\ &+ \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{gen}(\tau)] \right] p_{s+1,s+1}(\tau, t)(1 - p_{s+1,s+1}(\tau, t)) d\tau \\ &+ \int_0^t \left[ \frac{d}{d\tau} \text{Var}[X_{s+1}^{gen}(\tau)] \right] (p_{s+1,s+1}(\tau, t))^2 d\tau \end{aligned} \quad (B35)$$

$$E[Y_{s+1,k}(t)] = m_{s+1}(0)q_{s+1,s+1,k}(0, t) + \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{gen}(\tau)] \right] q_{s+1,s+1,k}(\tau, t) d\tau \quad (B36)$$

$$\begin{aligned} \text{Var}[Y_{s+1,k}(t)] &= m_{s+1}(0)q_{s+1,s+1,k}(0, t)(1 - q_{s+1,s+1,k}(0, t)) \\ &+ \int_0^t \left[ \frac{d}{d\tau} E[X_{s+1}^{gen}(\tau)] \right] q_{s+1,s+1,k}(\tau, t)(1 - q_{s+1,s+1,k}(\tau, t)) d\tau \end{aligned}$$

$$+ \int_0^t \left[ \frac{d}{d\tau} \text{var}[X_{s+1}^{\text{gen}}(\tau)] \right] (q_{s+1, s+1, k}(\tau, t))^2 d\tau \quad (\text{B37})$$

which are equations (23) through (26) in the text.

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Table 1. Transition probabilities obtained for bed 1 in numerical simulation.

Table 2. Transition probabilities obtained for bed 2 in numerical simulation.

Table 1. Transition probabilities obtained for bed 1 in numerical simulation.

$$p_{ij}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} e^{-0.07t} & 2.5(1-e^{-0.07t}) & 7.5(1-e^{-0.07t})-15e^{-0.06t} \\ 0 & e^{-0.05t} & 3(1-e^{-0.05t}) \\ 0 & 0 & e^{-0.06t} \end{array} \right] \end{matrix}$$

$$q_{ij1}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} 0.2857(1-e^{-0.07t}) & 0.2857+0.7143e^{-0.07t}-e^{-0.05t} & 0.1429-2.1429e^{-0.07t}-3e^{-0.05t}+5e^{-0.06t} \\ 0 & 0.4(1-e^{-0.05t}) & 0.2-1.2e^{-0.05t}+e^{-0.06t} \\ 0 & 0 & 0.3333(1-e^{-0.06t}) \end{array} \right] \end{matrix}$$

$$q_{ij2}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} 0 & 0 & 0.2858-4.2858e^{-0.07t}-6e^{-0.05t}+10e^{-0.06t} \\ 0 & 0 & 0.4-2.4e^{-0.05t}+2e^{-0.06t} \\ 0 & 0 & 0.6667(1-e^{-0.06t}) \end{array} \right] \end{matrix}$$

Table 2. Transition probabilities obtained for bed 2 in numerical simulation.

$$p_{ij}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} e^{-0.07t} & 2.5(1-e^{-0.07t}) & 7.5(1-e^{-0.07t})-15e^{-0.06t} \\ 0 & e^{-0.05t} & 3(1-e^{-0.05t}) \\ 0 & 0 & 0 \end{array} \right] \end{matrix}$$

$$q_{ij1}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} 0.2857(1-e^{-0.07t}) & 0.2857+0.7143e^{-0.07t}-e^{-0.05t} & 0.1429-2.1429e^{-0.07t}-3e^{-0.05t}+5e^{-0.06t} \\ 0 & 0.4(1-e^{-0.05t}) & 0.2-1.2e^{-0.05t}+e^{-0.06t} \\ 0 & 0 & 0 \end{array} \right] \end{matrix}$$

$$q_{ij2}(0, t) = \begin{matrix} & \begin{matrix} j \\ i \end{matrix} & \begin{matrix} 1 & 2 & 3 \end{matrix} \\ \begin{matrix} 1 \\ 2 \\ 3 \end{matrix} & \left[ \begin{array}{ccc} 0 & 0 & 0.2858-4.2858e^{-0.07t}-6e^{-0.05t}+10e^{-0.06t} \\ 0 & 0 & 0.4-2.4e^{-0.05t}+2e^{-0.06t} \\ 0 & 0 & 1 \end{array} \right] \end{matrix}$$

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Fig. 1. Schematic diagram of the general cell model system.

Fig. 2. Particle disintegration mechanism.

Fig. 3. Two-stage fluidized-bed operation considered in the numerical simulation.

Fig. 4. Transient mean and variance of size 1 particle population in bed 1 arising from initial contents:

$$\square = E[ X_1^1 ], * = \text{Var}[ X_1^1 ].$$

Fig. 5. Transient mean and variance of size 2 particle population in bed 1 arising from initial contents:

$$\square = E[ X_2^1 ], * = \text{Var}[ X_2^1 ].$$

Fig. 6. Transient mean and variance of size 3 particle population (in gms.) in bed 1 arising from initial contents:

$$\square = E[ X_3^1 ], * = \text{Var}[ X_3^1 ].$$

Fig. 7. Transient mean and variance of size 1 particle population arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ Y_{11}^1 ], * = \text{Var}[ Y_{11}^1 ].$$

Fig. 8. Transient mean and variance of size 2 particle population arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ Y_{21}^1 ], * = \text{Var}[ Y_{21}^1 ].$$

Fig. 9. Transient mean and variance of size 3 particle population (in gms.) arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ Y_{31}^1 ], * = \text{Var}[ Y_{31}^1 ].$$

Fig. 10. Transient mean and variance of size 3 particle population (in gms.) arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 2:

$$\square = E[ Y_{32}^1 ], * = \text{Var}[ Y_{32}^1 ].$$

Fig. 11. Transient mean and variance of size 1 particle population in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{11}^1 ], * = \text{Var}[ B_{11}^1 ].$$



Fig. 12. Transient mean and variance of size 2 particle population in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{12}^1 ], * = \text{Var}[ B_{12}^1 ].$$

Fig. 13. Transient mean and variance of size 3 particle population (in gms.) in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{13}^1 ], * = \text{Var}[ B_{13}^1 ].$$

Fig. 14. Transient mean and variance of size 1 particle population arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ B_{111}^1 ], * = \text{Var}[ B_{111}^1 ].$$

Fig. 15. Transient mean and variance of size 2 particle population arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ B_{121}^1 ], * = \text{Var}[ B_{121}^1 ].$$

Fig. 16. Transient mean and variance of size 3 particle population (in gms.) arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ B_{131}^1 ], * = \text{Var}[ B_{131}^1 ].$$

Fig. 17. Transient mean and variance of size 3 particle population (in gms.) arising from inflow to bed 1 and having exited from bed 1 through outlet stream 2:

$$\square = E[ B_{132}^1 ], * = \text{Var}[ B_{132}^1 ].$$

Fig. 18. Transient mean and variance of size 1 particle population in bed 2 arising from initial contents:

$$\square = E[ X_1^2 ], * = \text{Var}[ X_1^2 ].$$

Fig. 19. Transient mean and variance of size 2 particle population in bed 2 arising from initial contents:

$$\square = E[ X_2^2 ], * = \text{Var}[ X_2^2 ].$$

Fig. 20. Transient mean and variance of size 1 particle population arising from initial contents of bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ Y_{11}^2 ], * = \text{Var}[ Y_{11}^2 ].$$

Fig. 21. Transient mean and variance of size 2 particle population arising from initial contents of bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ Y_{21}^2 ], * = \text{Var}[ Y_{21}^2 ].$$

Fig. 22. Transient mean and variance of the total size 3 particle population (in gms.) in bed 2 arising from initial contents of bed 2 and having exited through outlet stream 2:

$$\square = E[ Y_{32}^2 ], * = \text{Var}[ Y_{32}^2 ].$$

Fig. 23. Transient mean and variance of size 1 particle population in bed 2 arising from inflow to bed 2:

$$\square = E[ B_{11}^2 ], * = \text{Var}[ B_{11}^2 ].$$

Fig. 24. Transient mean and variance of size 2 particle population in bed 2 arising from inflow to bed 2:

$$\square = E[ B_{12}^2 ], * = \text{Var}[ B_{12}^2 ].$$

Fig. 25. Transient mean and variance of size 1 particle population arising from inflow to bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ B_{111}^2 ], * = \text{Var}[ B_{111}^2 ].$$

Fig. 26. Transient mean and variance of size 2 particle population arising from inflow to bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ B_{121}^2 ], * = \text{Var}[ B_{121}^2 ].$$

Fig. 27. Transient mean and variance of the total size 3 particle population (in gms.) in bed 2 arising from inflow to bed 2 and having exited from bed 2 through outlet stream 2:

$$\square = E[ B_{132}^2 ], * = \text{Var}[ B_{132}^2 ].$$

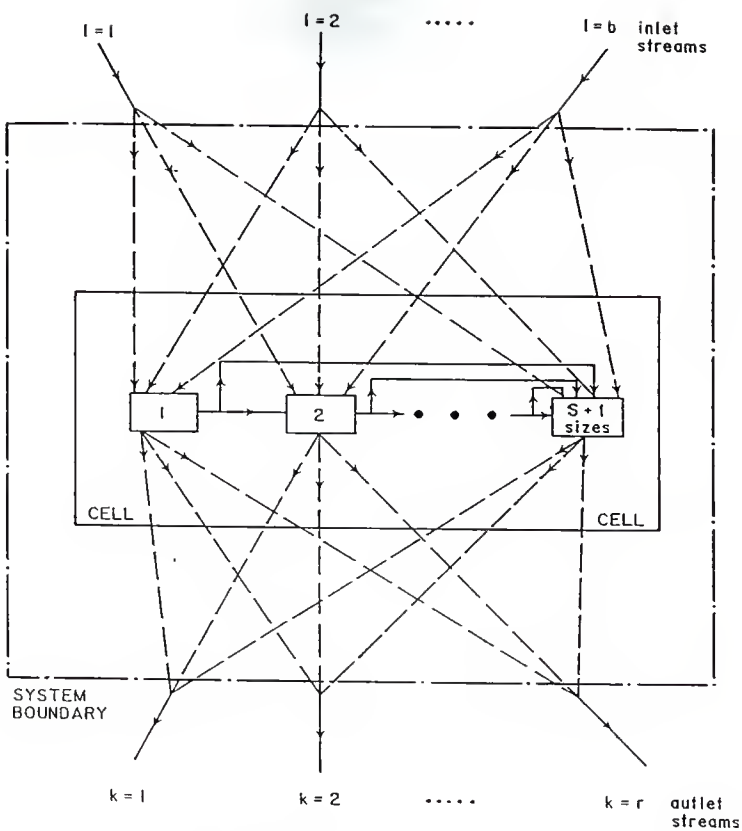


Fig. 1. Schematic diagram of the general cell model system.

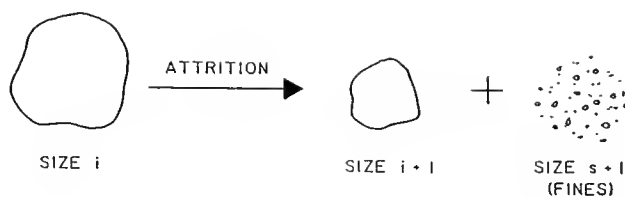


Fig. 2. Particle disintegration mechanism.

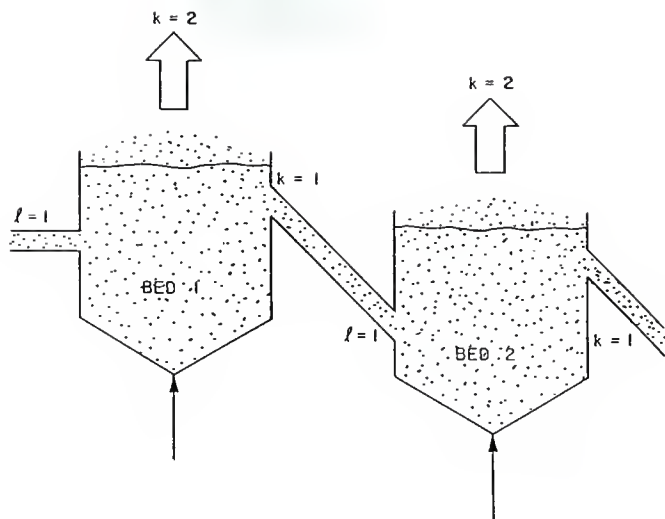


Fig. 3. Two-stage fluidized-bed operation considered in the numerical simulation.

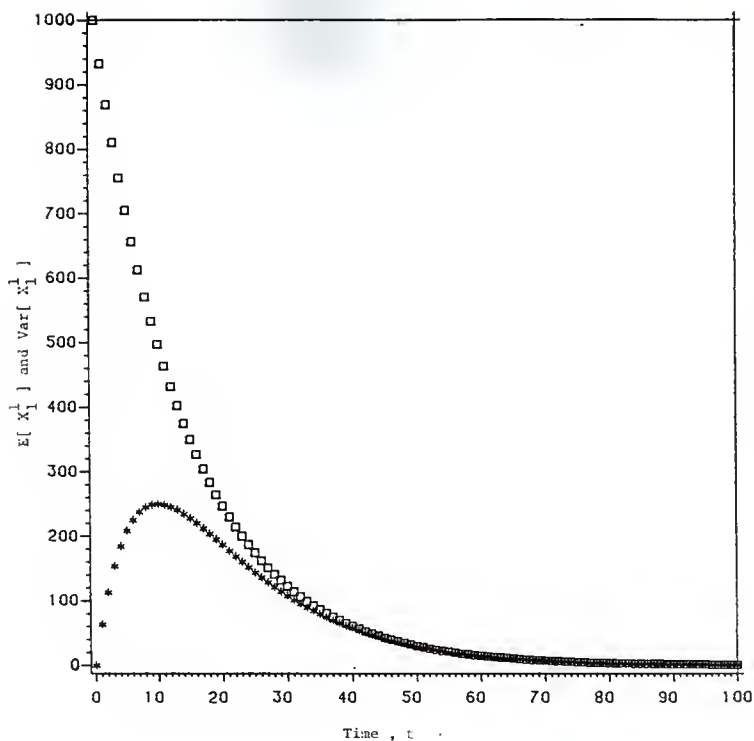


Fig. 4. Transient mean and variance of size 1 particle population in bed 1 arising from initial contents:

$$\square = E[X_1^1], \quad * = \text{Var}[X_1^1].$$

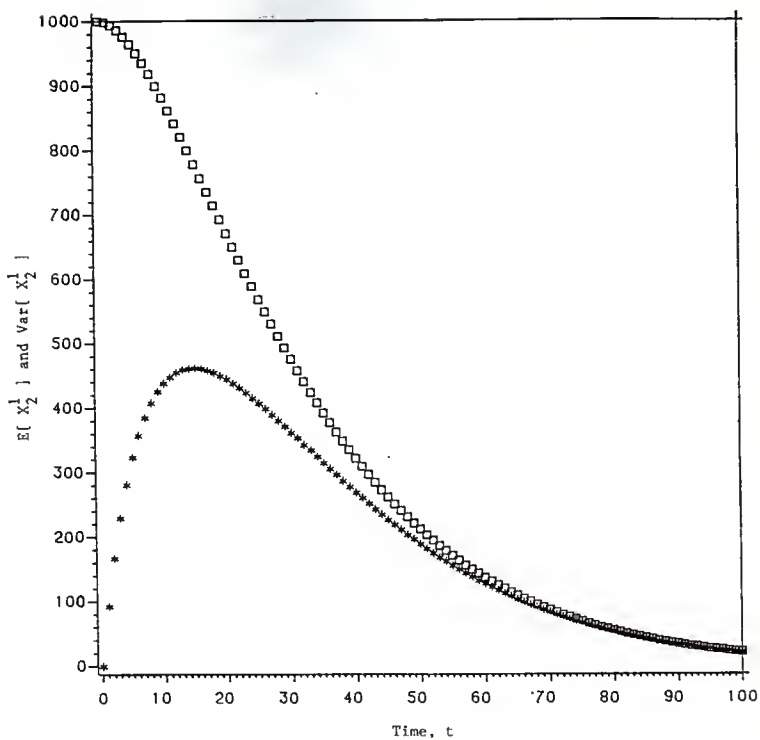


Fig. 5. Transient mean and variance of size 2 particle population in bed 1 arising from initial contents:  
 $\square = E[ X_2^1 ]$ ,  $* = Var[ X_2^1 ]$ .

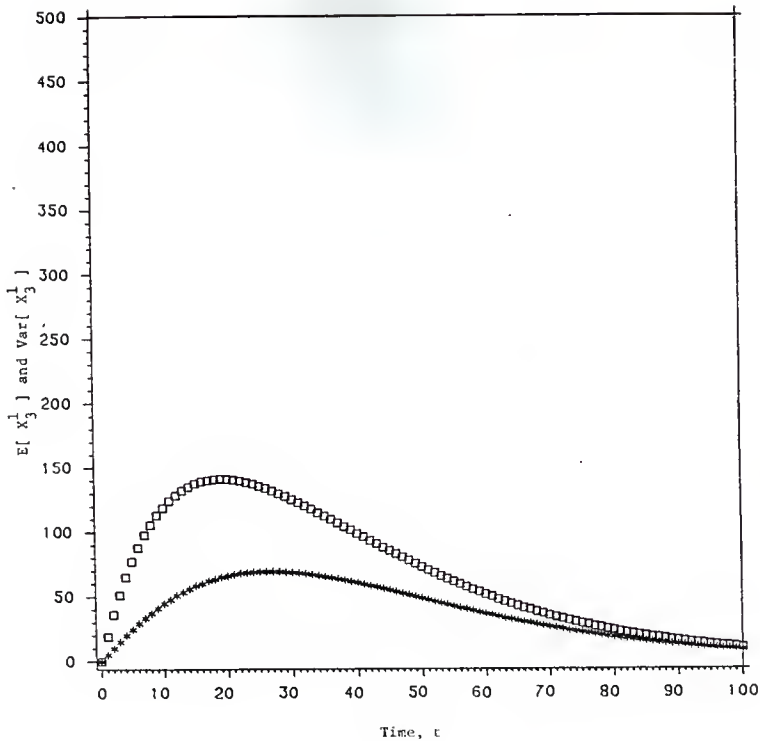


Fig. 6. Transient mean and variance of size 3 particle population (in gms.) in bed 1 arising from initial contents:

$$\square = E[X_3^1], * = Var[X_3^1].$$



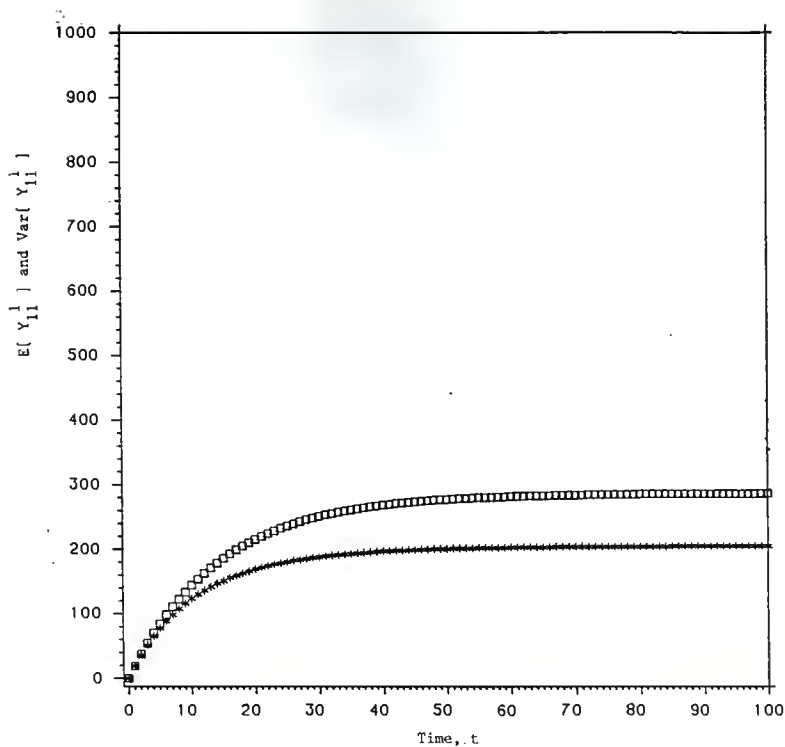


Fig. 7. Transient mean and variance of size 1 particle population arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ Y_{11}^1 ], \quad * = Var[ Y_{11}^1 ].$$

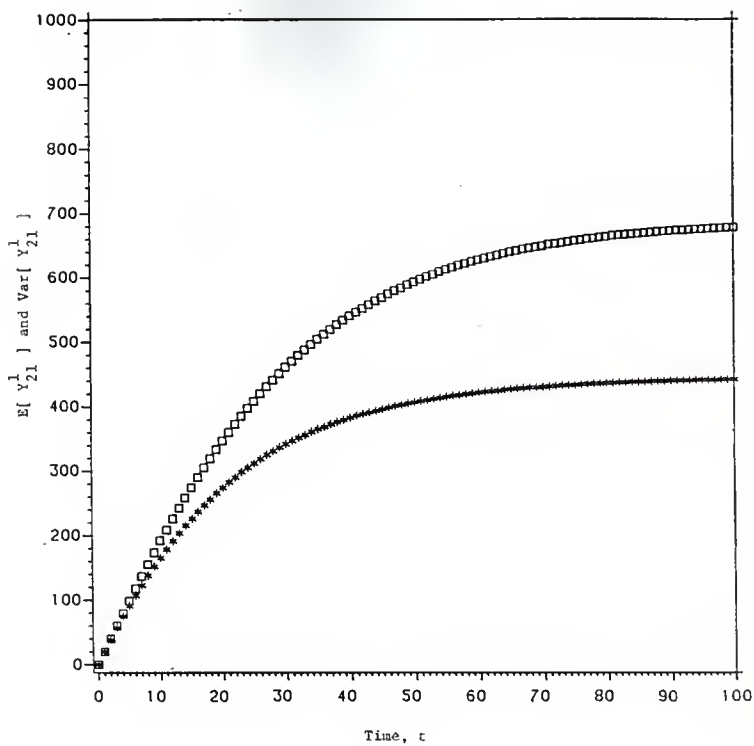


Fig. 8. Transient mean and variance of size 2 particle population arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E\{v_{21}^1\}, \quad * = \text{Var}\{v_{21}^1\}.$$

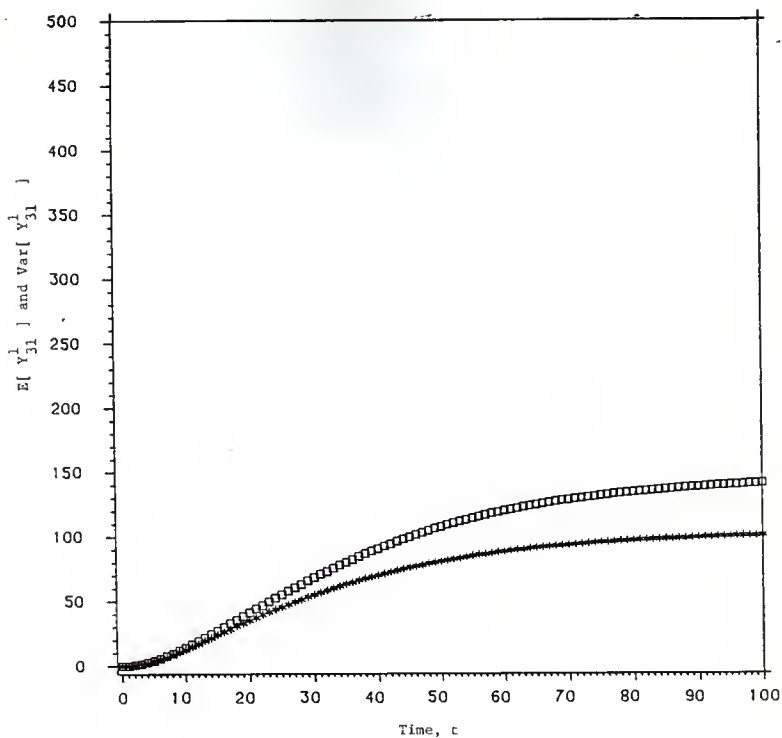


Fig. 9. Transient mean and variance of size 3 particle population (in gms.) arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[ Y_{31}^1 ], * = Var[ Y_{31}^1 ].$$

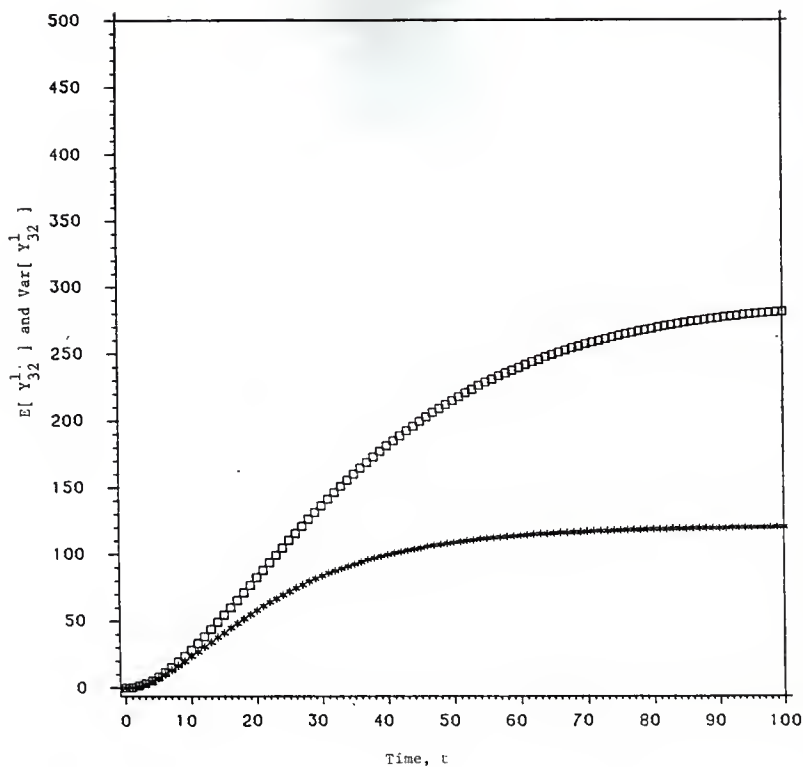


Fig. 10. Transient mean and variance of size 3 particle population (in gms.) arising from initial contents of bed 1 and having exited from bed 1 through outlet stream 2:

$$\square = E[ y_{32}^1 ], * = Var[ y_{32}^1 ].$$

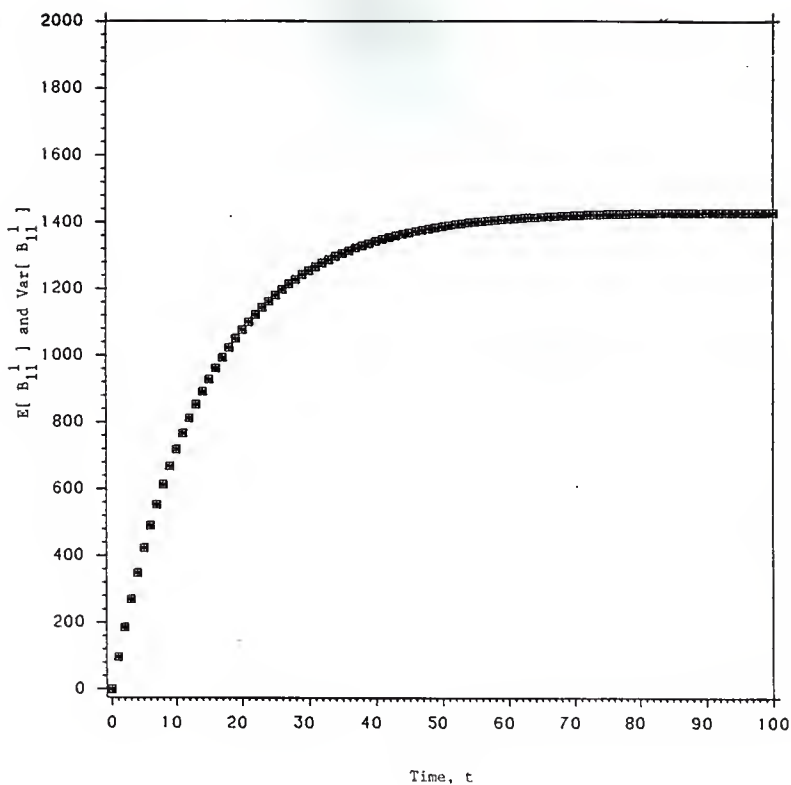


Fig. 11. Transient mean and variance of size 1 particle population in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{11}^1 ], * = \text{Var}[ B_{11}^1 ].$$

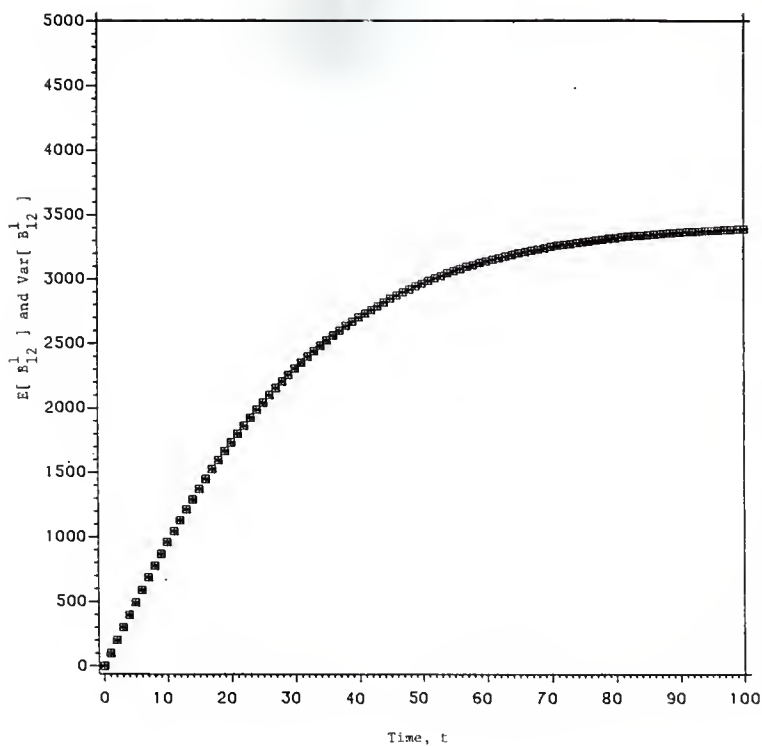


Fig. 12. Transient mean and variance of size 2 particle population in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{12}^1 ], * = \text{Var}[ B_{12}^1 ].$$

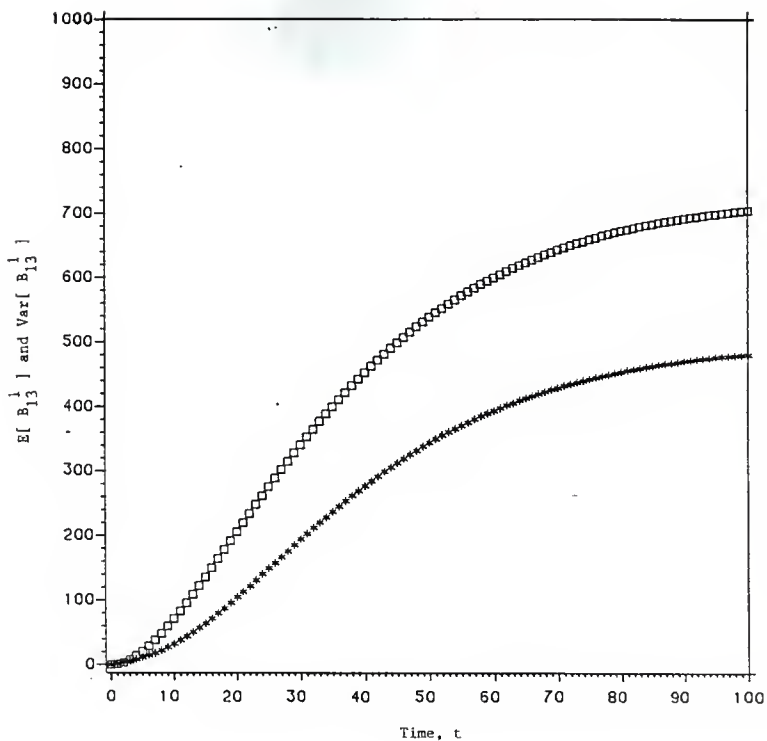


Fig. 13. Transient mean and variance of size 3 particle population (in gms.) in bed 1 arising from inflow to bed 1:

$$\square = E[ B_{13}^1 ], * = \text{Var}[ B_{13}^1 ].$$

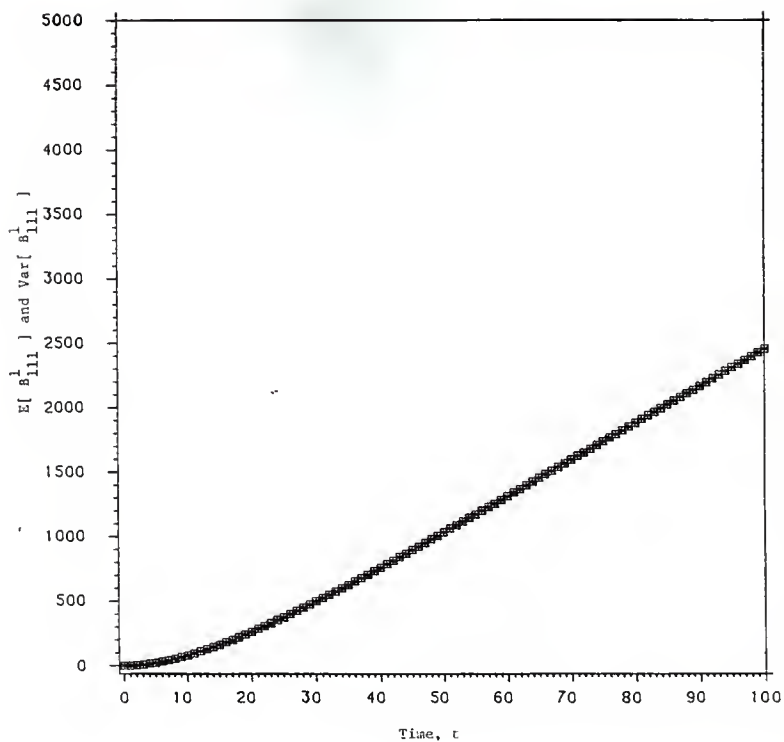


Fig. 14. Transient mean and variance of size 1 particle population arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E[B_{111}^1], * = Var[B_{111}^1].$$



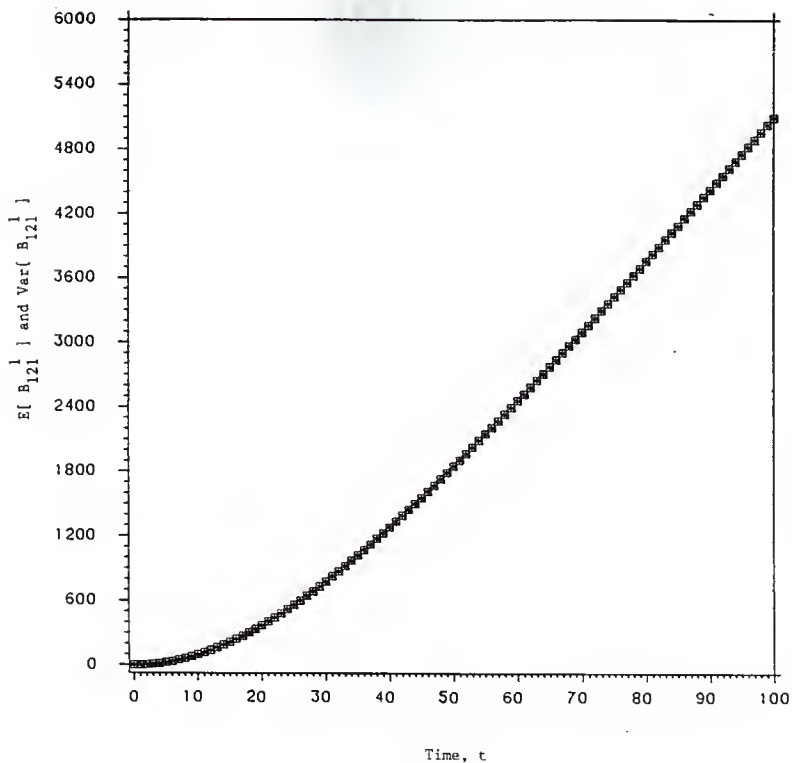


Fig. 15. Transient mean and variance of size 2 particle population arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E\{B_{121}^1\}, * = \text{Var}\{B_{121}^1\}.$$

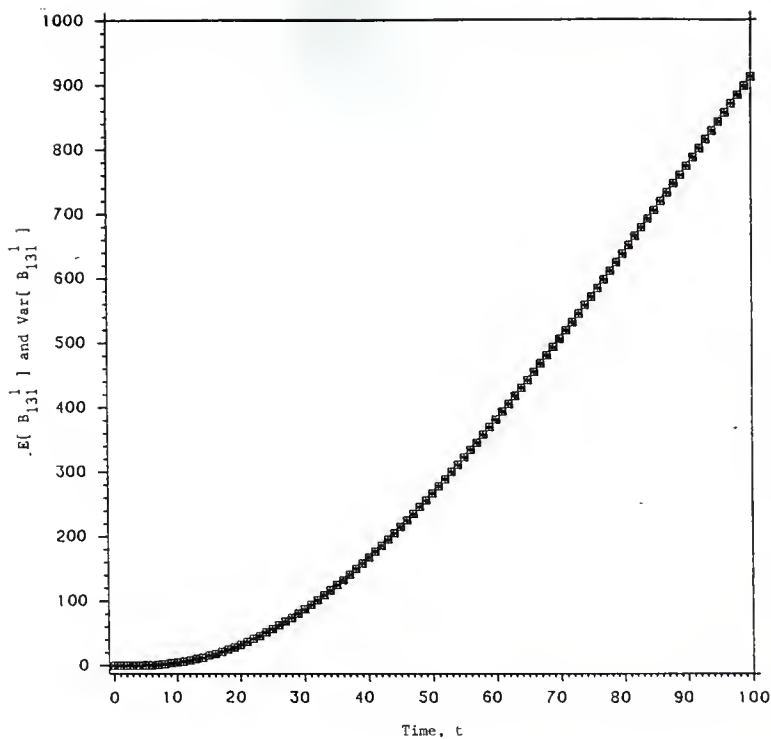


Fig. 16. Transient mean and variance of size 3 particle population (in gms.) arising from inflow to bed 1 and having exited from bed 1 through outlet stream 1:

$$\square = E\{ B_{131}^1 \}, * = \text{Var}\{ B_{131}^1 \}.$$

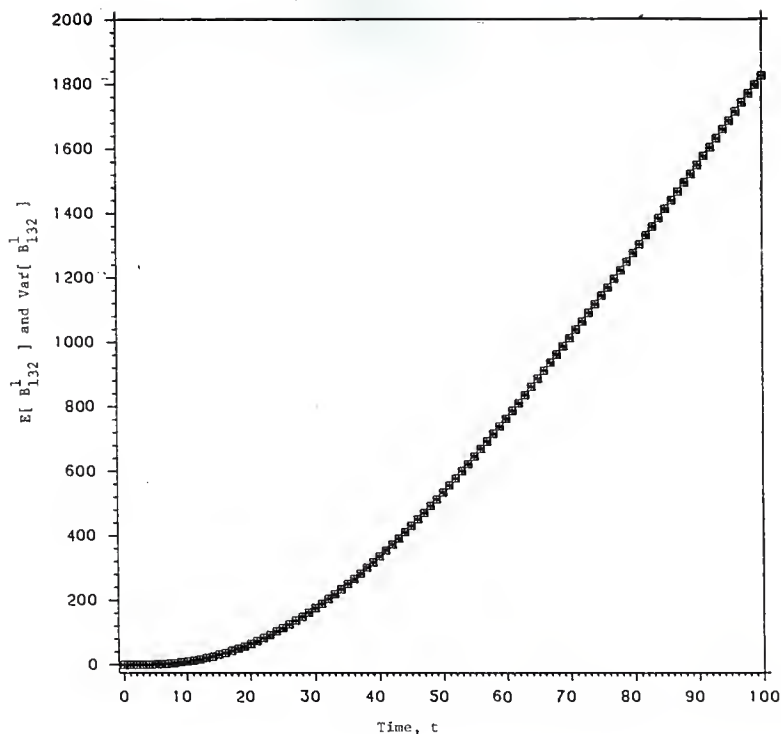


Fig. 17. Transient mean and variance of size 3 particle population (in gms.) arising from inflow to bed 1 and having exited from bed 1 through outlet stream 2:

$$\square = E[B_{132}^1], \quad * = \text{Var}[B_{132}^1].$$

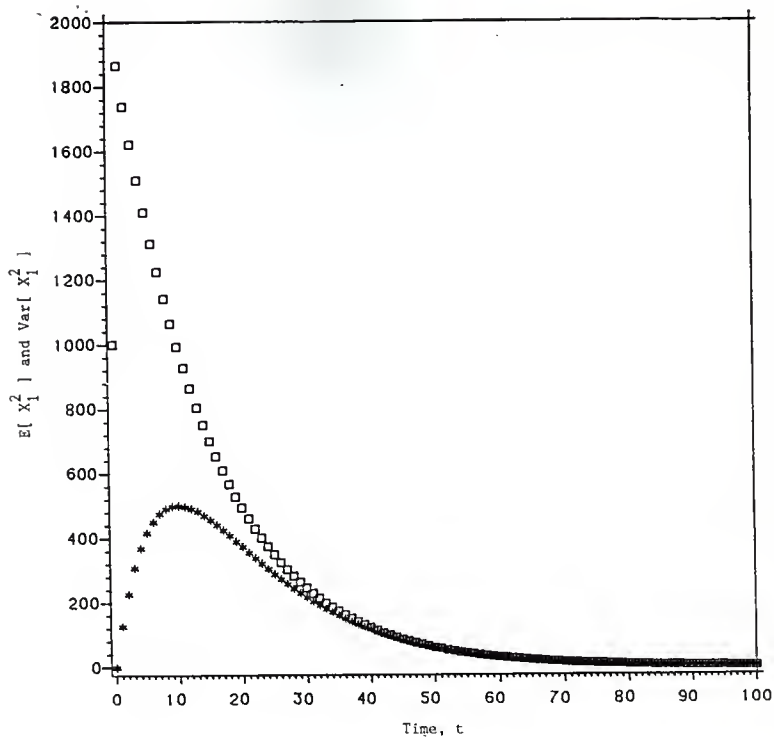


Fig. 18. Transient mean and variance of size 1 particle population in bed 2 arising from initial contents:

$$\square = E[ X_1^2 ], * = \text{Var}[ X_1^2 ].$$

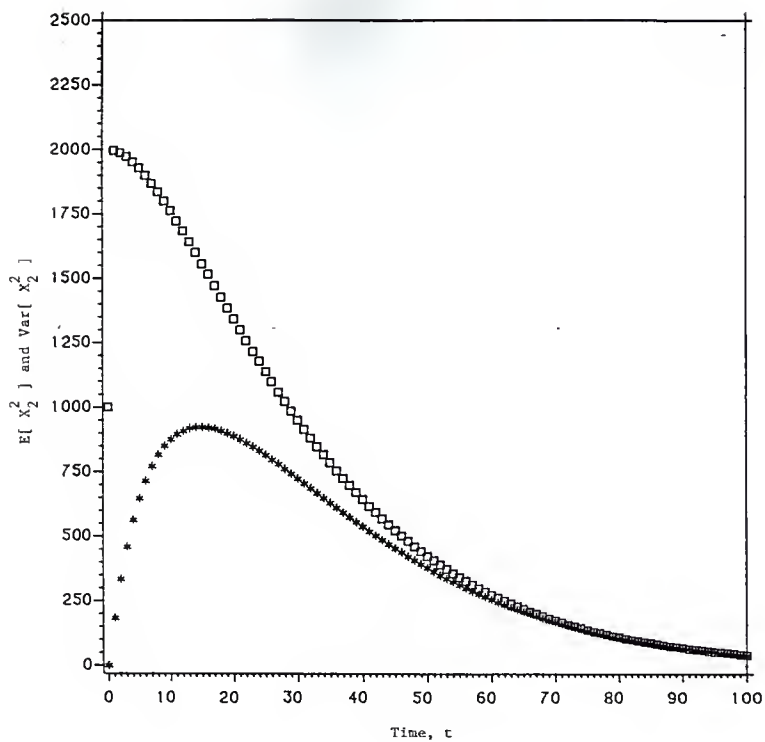


Fig. 19. Transient mean and variance of size 2 particle population in bed 2 arising from initial contents:

$$\square = E[ X_2^2 ], * = \text{Var}[ X_2^2 ].$$

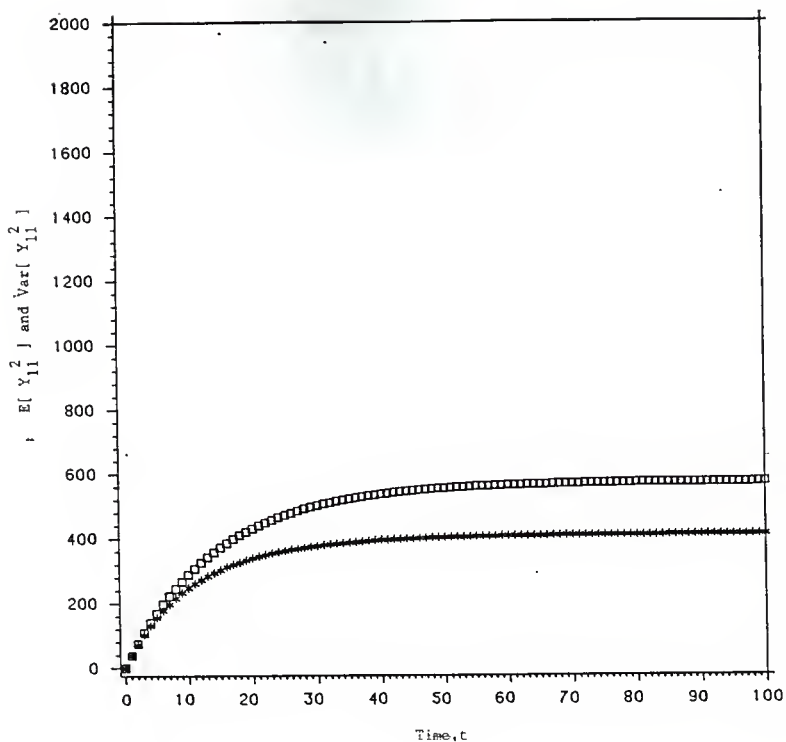


Fig. 20. Transient mean and variance of size 1 particle population arising from initial contents of bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ Y_{11}^2 ], * = Var[ Y_{11}^2 ].$$

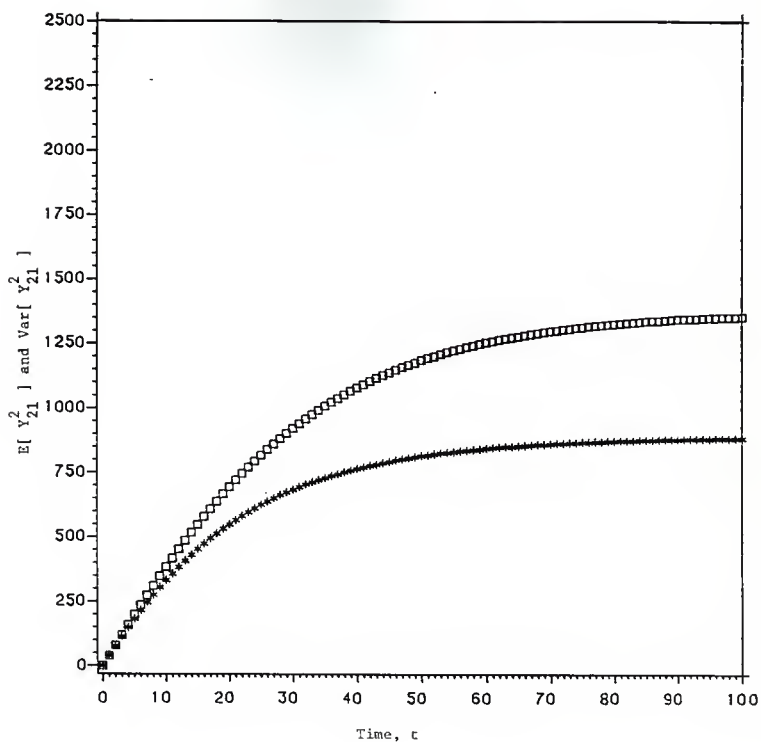


Fig. 21. Transient mean and variance of size 2 particle population arising from initial contents of bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[y_{21}^2], \quad * = \text{Var}[y_{21}^2].$$

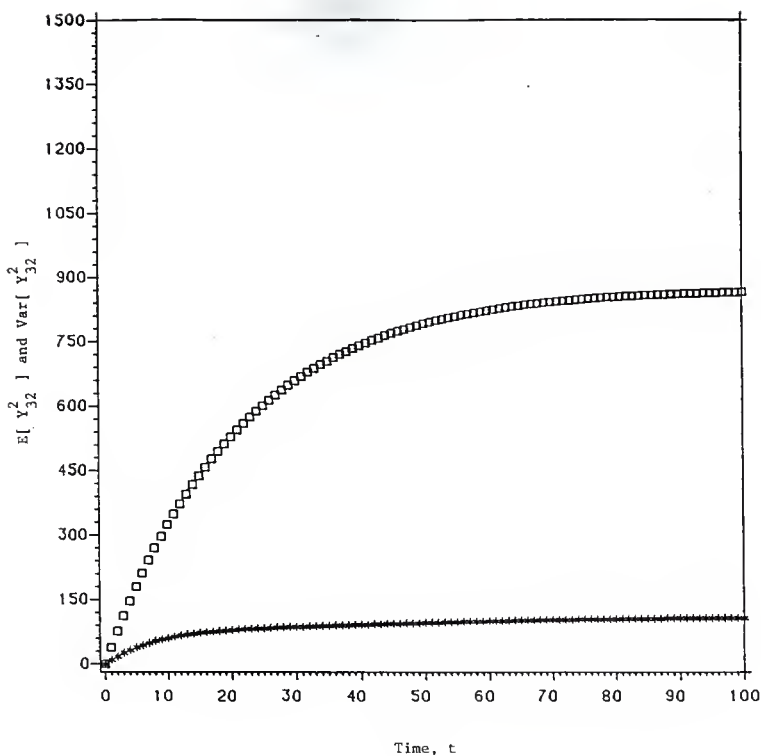


Fig. 22. Transient mean and variance of the total size 3 particle population (in gms.) in bed 2 arising from initial contents of bed 2 and having exited through outlet stream 2:

$$\square = E[Y_{32}^2], * = \text{Var}[Y_{32}^2].$$



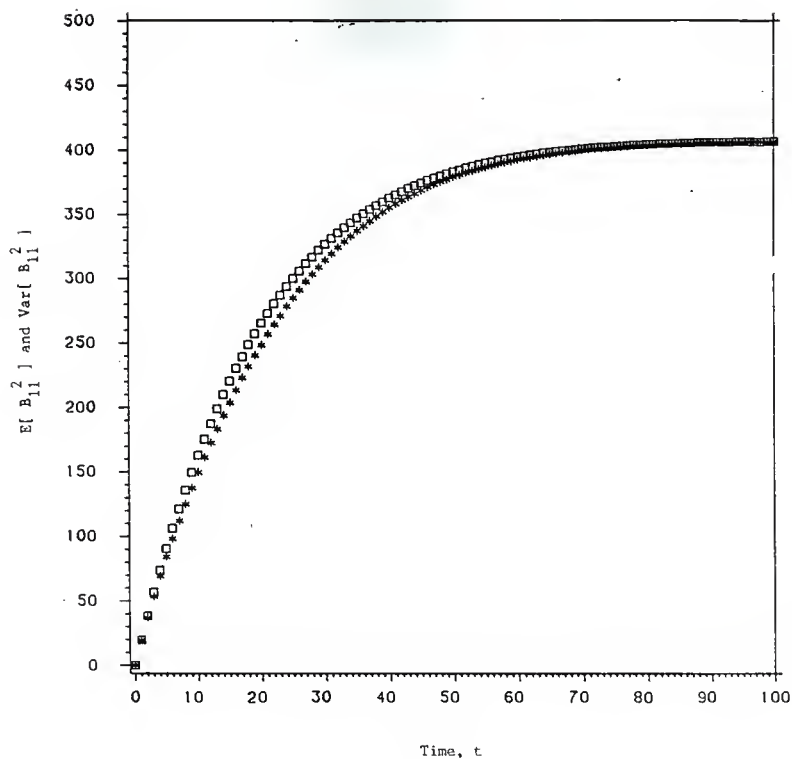


Fig. 23. Transient mean and variance of size 1 particle population in bed 2 arising from inflow to bed 2:

$$\square = E[ B_{11}^2 ], * = \text{Var}[ B_{11}^2 ].$$

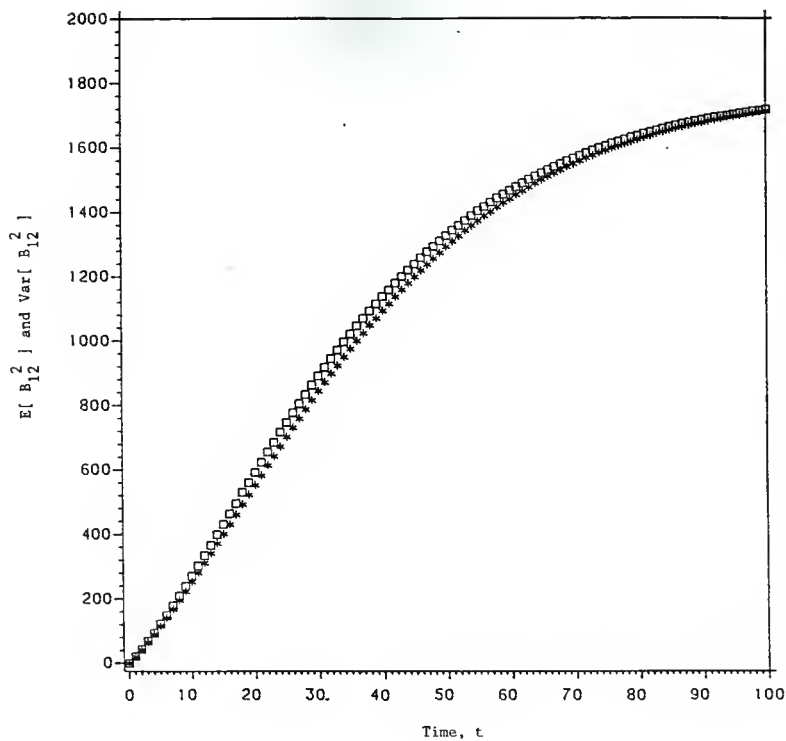


Fig. 24. Transient mean and variance of size 2 particle population in bed 2 arising from inflow to bed 2:

$$\square = E[B_{12}^2], * = \text{Var}[B_{12}^2].$$

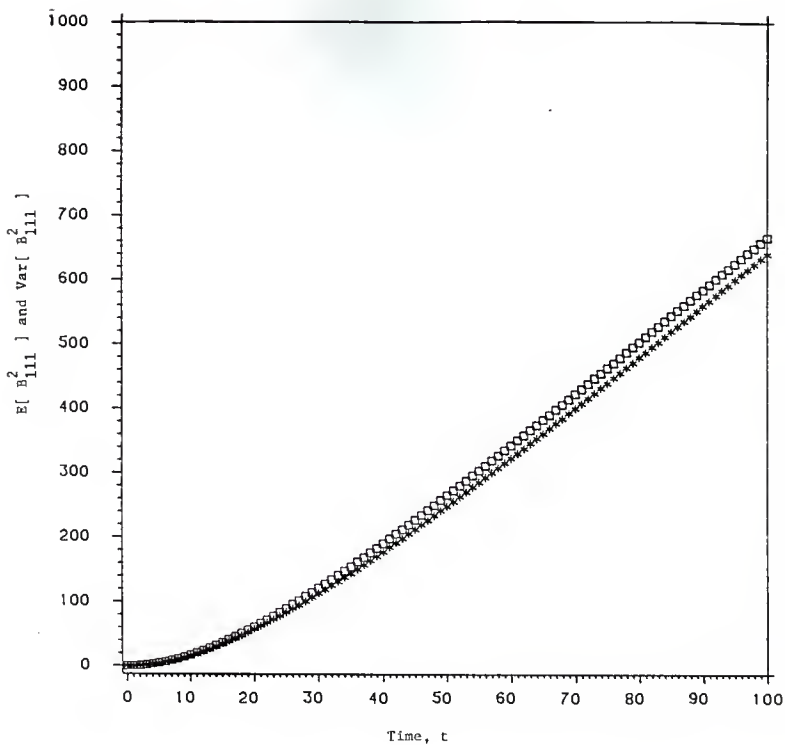


Fig. 25. Transient mean and variance of size 1 particle population arising from inflow to bed 2 and having exited from bed 2 through outlet stream 1:

$$\mu = E[B_{111}^2], \quad \sigma^2 = \text{Var}[B_{111}^2].$$

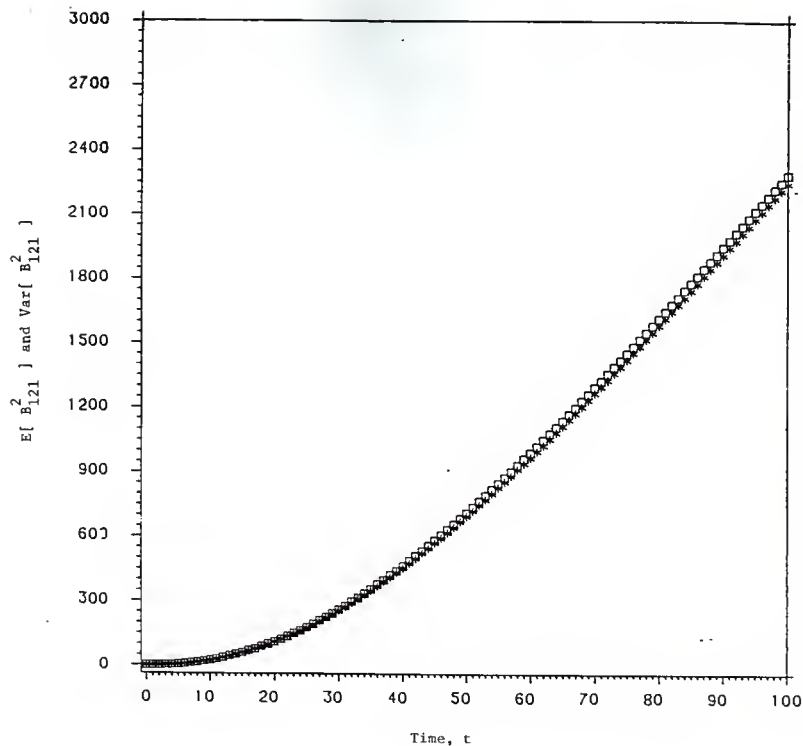


Fig. 26. Transient mean and variance of size 2 particle population arising from inflow to bed 2 and having exited from bed 2 through outlet stream 1:

$$\square = E[ B_{121}^2 ], * = \text{Var}[ B_{121}^2 ].$$

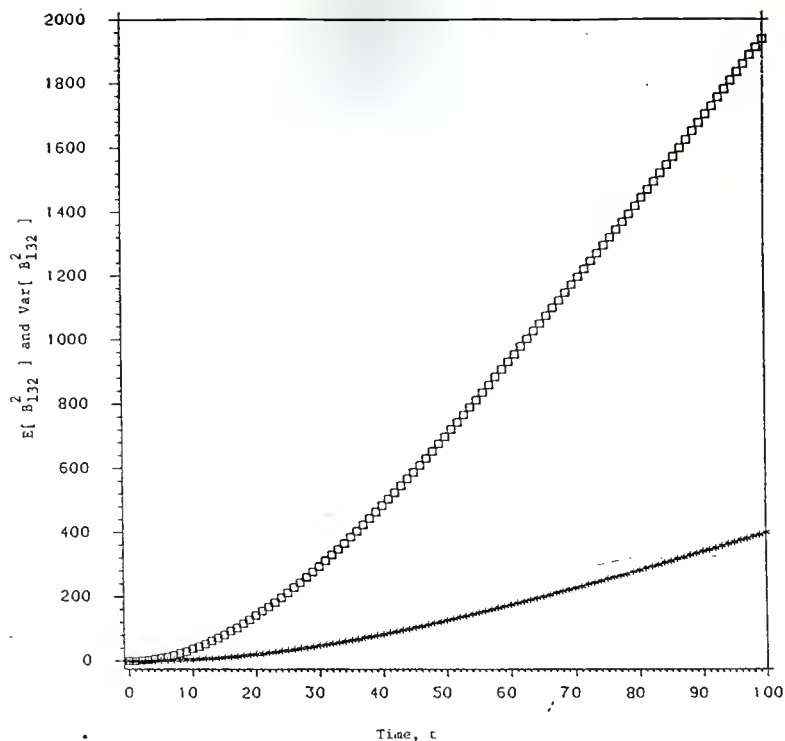


Fig. 27. Transient mean and variance of the total size 3 particle population (in gms.) in bed 2 arising from inflow to bed 2 and having exited from bed 2 through outlet stream 2:

$$\square = [ B_{132}^2 ], * = \text{Var}[ B_{132}^2 ].$$

## CHAPTER 3

### STOCHASTIC MODELING OF NON-LINEAR SIEVING KINETICS

#### INTROOUCION

Sieving plays a predominant role in process engineering involving particulate systems as a means for particle size analysis or separation (see, e.g., Gupta et al., 1975). Sieving usually refers to batch operations, while screening refers to continuous operations in practice, although the principles of size separation in both modes of operation remain essentially identical.

The accuracy of the standard specifications for sieve analysis depends on the available information on sieving. Oespite its popularity and prominence, the kinetics of the sieving process have received relatively little attention, much of the investigations having been conducted in recent years (see, e.g., Miwa, 1974).

Simply stated, the sieving process involves repeated encounters between particles and the sieve surface which may result in passage through, blinding (also referred to as clogging) or retention on the surface of the sieve. It is often stated that the process is indeed random, and attempts have been made at describing its kinetics in the terminal stages of sieving through a simple stochastic model (see, e.g., Kapur et al., 1977). The model is based on the first-order kinetics law, obtained from simple probabilistic considerations.

According to Kaye (1962), the probability that any of the particles on the sieve will pass through a sieve mesh depends on:

1. the physical characteristics of the particles,
2. the number density of the particles,
3. the size distribution of the particles,
4. the characteristic motion of the sieve, and
5. the geometry of the sieve mesh, and the size distribution of the mesh apertures.

A complete mathematical description of the process is evasive due to the complexity arising from the blending of these factors.

The feed to the sieve can be broadly classified as oversize particles, incapable of passing through the sieve, undersize particles, capable of readily passing through the sieve, and near mesh size particles whose sizes vary around the aperture size; these near mesh size particles can blind apertures. When particulate mixtures are classified by size or separated by sieving, proper classification or separation is ensured by the passage of the near mesh size particles. Hence, it is essential that the kinetics of the passage of these particles through the sieve be studied.

In the present work, a general rate equation or model for the passage of near mesh size particles through the apertures of a sieve in the presence of blinding and in the presence of oversize particles on the sieve is derived based on stochastic theory. The resultant equation describes the entire sieving operation. The model provides a measure of the inherent significant fluctuations with respect to the particle size distributions and determines the manner in which the near mesh size particles affect the weight fraction passing through the sieve.

Various expressions have been proposed to identify the near mesh size particles with respect to a given aperture or sieve opening size (see, e.g., Whitby, 1958). Whitby (1958) has confirmed that the passage of these particles is clearly stochastic rather than deterministic; it does not depend strictly on the ratio of the particle size to the aperture opening size. Nevertheless, Rendell (1964) has proposed a deterministic relationship for the rate of flow of particles through the sieve based on the ratio of the particle size,  $d_p$ , to the aperture opening size,  $D_a$ ; it is expressed as

$$R \propto \left( \frac{D_a}{d_p} - k \right)^m \quad (1)$$

where  $k$  is a constant ranging from 0.8 to 1.7, and  $m$  a constant ranging from 1.5 to 2.0.

Clogging of the apertures or sieve blinding occurs almost universally in sieving operations with near mesh size particles; it limits the passage of particles. The present model considers clogging as a birth-death process and includes its effect on the flow of particles through the sieve mesh in the presence of oversize particles. By considering the particle population on the sieve to possess the Markov property, a bivariate master equation is formulated to obtain the rate of passage of the near mesh size particles through the sieve and the rate of the concomitant sieve blinding. The master equation does not yield an explicit analytical solution and hence a rational approximation technique, the system size expansion is utilized to solve the master equation. Analytical solutions are obtained for the master equation



describing the sieving operation in the case of no oversize particles being present on the sieve.

# DERIVATION OF THE BIVARIATE MASTER EQUATION

Let the random variable  $N$  denote the number of near mesh size particles actively present (not lodged in the apertures) on the sieve surface,  $A_c$  the number of clogged apertures, and  $n_{os}$  the number (specified) of oversize particles present on the sieve. Unless explicitly stated otherwise, any reference to particles henceforth implies the near mesh size particles. The joint probability of the two random variables,  $N$  and  $A_c$ , will be denoted as  $P(n, a_c; t)$ . The state space of  $N$  consists of the domain of integers,  $\{n \in (0, 1, 2, \dots, n_0)\}$ , with  $n_0$  corresponding to the initial number of near mesh size particles on the sieve. The state space of  $A_c$  also consists of the domain of integers,  $\{a_c \in (0, 1, 2, \dots, a_0)\}$ , with  $a_0$  corresponding to the total number of sieve openings.  $P(n, a_c; t)$  is interpreted as  $P(N=n, A_c=a_c; t)$  or the joint probability that the random variable  $N$  has a value of  $n$  and the random variable  $A_c$  has a value of  $a_c$  at time  $t$ . The conditional probability  $P(n, a_c; t | n', a'_c; \tau)$  is the probability that the random variables  $N$  and  $A_c$  assume the values of  $n$  and  $a_c$ , respectively, at time  $t$ , given that they had values of  $n'$  and  $a'_c$ , respectively, at time  $\tau$ .

## Probability Balance

Assuming that the states of the two populations possess the Markov property,  $P(n, a_c; t)$  is expressed as (see, e.g., Kapur et al., 1977)

$$P(n_t, a_{c,t}; t) = \sum_{n', a'_c} P(n, a_c; t | n', a'_c; \tau) P(n', a'_c; \tau) \quad (2)$$

For the system under consideration, the following conditional probabilities are defined:

$$P(n-1, a_c; t+\Delta t | n, a_c; t) = \kappa_{n, a_c} \Delta t + o(\Delta t) \quad (3)$$

$$P(n, a_{c, t}^{-1}; t+\Delta t | n, a_c; t) = \mu_{a_c} \Delta t + o(\Delta t) \quad (4)$$

$$P(n-1, a_c+1; t+\Delta t | n, a_c; t) = \lambda_{n, a_c} \Delta t + o(\Delta t) \quad (5)$$

$$P(n, a_c; t+\Delta t | n, a_c; t) = 1 - \kappa_{n, a_c} \Delta t - \mu_{a_c} \Delta t - \lambda_{n, a_c} \Delta t + o(\Delta t) \quad (6)$$

The probability of more than one event, either a passage, clogging or unclogging, occurring in the time interval  $(t, t+\Delta t)$ , is given by  $o(\Delta t)$ , where  $o(\Delta t)$  is an arbitrary function of  $\Delta t$  such that

$$\lim_{\Delta t \rightarrow 0} \frac{o(\Delta t)}{\Delta t} = 0$$

Equation (3) states that the transition probability of a near mesh size particle passing through the sieve in the time interval  $(t, t+\Delta t)$  is  $\kappa_{n, a_c} \Delta t + o(\Delta t)$  where  $\kappa_{n, a_c}$  is a function of  $n$  as well as  $a_c$ , as will be elaborated later. Equation (4) relates to the loss or death of a clogged aperture, i.e., the transition probability of a clogged aperture freeing itself (by the passage of the near mesh size particle blinding that aperture, through the sieve mesh) in the time interval  $(t, t+\Delta t)$  is  $\mu_{a_c} \Delta t + o(\Delta t)$  where  $\mu_{a_c}$  is a function of  $a_c$ . The particle blinding an aperture is assumed to pass through the sieve, forced by the other particles on the sieve. Equation (5) states that the transition probability of the appearance or birth of a clogged aperture, i.e., an aperture being clogged in the time interval  $(t, t+\Delta t)$ , concomitant with a

decrease in the population size  $n$  by 1, is  $\lambda_{n,a_c} \Delta t + o(\Delta t)$  where  $\lambda_{n,a_c}$  is a function of  $a_c$  and  $n$ . Equation (6) relates to the probability of status quo, stating that the probability of no change in the system in the time interval  $(t, t+\Delta t)$  is

$$1 - \kappa_{n,a_c} \Delta t - \mu_{a_c} \Delta t - \lambda_{n,a_c} \Delta t - o(\Delta t)$$

A probability balance is obtained by considering the various events that can occur in a small time interval  $\Delta t$ . It is assumed that any one of the above events occurs in this time interval  $\Delta t$ . The probability of  $n$  particles remaining on the sieve and  $a_c$  clogged apertures being present at time  $t+\Delta t$  is the sum of the following:

1. the probability of  $n+1$  particles on the sieve and  $a_c$  clogged apertures present at time  $t$ , multiplied by the probability of a particle passing through the sieve during the time interval  $\Delta t$ ,
2. the probability of  $n$  particles on the sieve and  $a_c + 1$  clogged apertures present at time  $t$ , multiplied by the probability of a clogged aperture freeing itself by the passage of the clogging particle through the sieve during the time interval  $\Delta t$ ,
3. the probability of  $n+1$  particles on the sieve and  $a_c - 1$  clogged apertures present at time  $t$ , multiplied by the probability of a particle on the sieve clogging on aperture during the time interval  $\Delta t$ , and

4. the probability of  $n$  particles on the sieve and  $a_c$  clogged apertures present at time  $t$ , multiplied by the probability of no change in states of  $n$  and  $a_c$  during the time interval  $\Delta t$ .

This gives rise to

$$\begin{aligned}
 & P(n_t, a_{c,t}; t + \Delta t) \\
 &= P(n+1, a_c; t) [\kappa_{n+1, a_c} \Delta t + o(\Delta t)] \\
 &\quad + P(n, a_c + 1; t) [\mu_{a_c + 1} \Delta t + o(\Delta t)] \\
 &\quad + P(n+1, a_c - 1; t) [\lambda_{n+1, a_c - 1} \Delta t + o(\Delta t)] \\
 &\quad + P(n, a_c; t) [1 - \kappa_{n, a_c} \Delta t - \mu_{a_c} \Delta t - \lambda_{n, a_c} \Delta t - o(\Delta t)] \quad (7)
 \end{aligned}$$

Rearranging this expression, dividing throughout by  $\Delta t$  and taking the limit as  $\Delta t \rightarrow 0$  give rise to the bivariate master equation

$$\begin{aligned}
 \frac{dP(n, a_c; t)}{dt} &= (E_n - 1) \kappa_{n, a_c} P(n, a_c; t) \\
 &\quad + (E_{a_c} - 1) \mu_{a_c} P(n, a_c; t) \\
 &\quad + (E_{a_c}^{-1} E_n - 1) \lambda_{n, a_c} P(n, a_c; t) \quad (8)
 \end{aligned}$$

where  $E$  is the one-step operator, defined by its effect on an arbitrary function  $f(n)$  as (see, e.g., van Kampen, 1958)

$$E f(n) = f(n+1) \text{ and } E^{-1} f(n) = f(n-1)$$

The expressions for the transition intensities for the system under study are derived as follows:

The particulate system comprises  $n$  near mesh size particles out of a total of  $(n_{os} + n)$  particles on the sieve; all these particles are

assumed to have equal access to the  $(a_o - a_c)$  open apertures. To pass through or blind an aperture, a particle first has to gain access to a sieve opening. This problem is akin to that of picking a ball from a box of black and white balls and deriving the probability of this event resulting in an outcome of a white ball. Hence, the probability of passage of a particle through an aperture can be envisioned as the probability of a particle, from a total of  $n_{os} + n$  particles actively present on the sieve, gaining access to a sieve opening, multiplied by the probability of passage for this particle (assumed to be identical for all of the near mesh size particles) through the opening. The transition intensity then includes the number of near mesh size particles, oversize particles and the fraction of apertures that are open; it is expressed as

$$\kappa_{n,a_c} = \frac{\kappa n}{(n_{os} + n)} \frac{(a_o - a_c)}{a_o}$$

The value of  $\kappa$  will depend on the physical properties of the material being sieved, characteristics of the sieve motion and sieve characteristics such as the shape of the apertures. Some effects of these factors have been studied (see e.g., Whitby, 1958). The apertures are assumed to be similar in size such that the probability of passage of a particle through an aperture is the same for all apertures. It is further assumed that  $\kappa$  does not depend on the number of undersize particles present on the sieve, i.e., the probability of passage of a near mesh size particle at any instant is not affected by an undersize particle either being present or passing through an aperture. The size of the

system considered will correspond to a normal loading of several layers. It is assumed that the sieve motion accomplishes the free movement of the particles in all directions. The intensity of an aperture being clogged  $\lambda_{n,a_c}$  is derived by the same token as  $\kappa_{n,a_c}$ ; it is expressed as

$$\lambda_{n,a_c} = \frac{\lambda n}{(n_{os} + n)} \frac{(a_o - a_c)}{a_o}$$

Its validity is conditional on the number of particles being larger than  $(a_o - a_c)$  (with, initially, more than a single layer of particles present). This is because a different mechanism prevails when

$$(n_{os} + n) < (a_o - a_c)$$

Also, each aperture has a certain probability of being unclogged; it is assumed that a particle frees itself by passing through the sieve. The intensity of unclogging is then expressed as

$$\mu_{a_c} = \frac{\mu a_c}{a_o}$$

On substituting the corresponding expressions for the intensities of transition, the master equation assumes the form

$$\begin{aligned} \frac{dP(n, a_c; t)}{dt} = & (E_n - 1) \frac{\kappa n}{(n_{os} + n)} \frac{(a_o - a_c)}{a_o} P(n, a_c; t) \\ & + (E_{a_c} - 1) \frac{\mu a_c}{a_o} P(n, a_c; t) \\ & + (E_{a_c}^{-1} E_n - 1) \frac{\lambda n}{(n_{os} + n)} \frac{(a_o - a_c)}{a_o} P(n, a_c; t) \end{aligned} \quad (9)$$

### System Size Expansion

The bivariate master equation, as expressed by equation (9), is not amenable to an explicit analytical solution for the probability joint distribution; hence, an approximation procedure is developed to solve the above equation. The system size expansion technique (see, e.g., van Kampen, 1958) can be employed here, since the changes in the magnitudes of random variables,  $N$  and  $A_C$ , in the time interval considered are sufficiently small compared to the value of the random variables themselves. In adopting the system size expansion, the joint probability distribution  $P(n, a_C; t)$  is expected to have a sharp peak at some position of order  $\Omega$ , the system size parameter, corresponding to the macroscopic value while its width will be of the order of  $\Omega^{1/2}$  representing the fluctuation (see, e.g., van Kampen, 1981). The expansion parameter  $\Omega$  governs the extent of fluctuations. Often,  $\Omega$  can simply be the size of the system. Here,  $a_0$  is chosen as the system size parameter  $\Omega$ ;  $n_0$  is considered to be of the same order of magnitude as  $a_0$ , and therefore, it is expressed in terms of  $a_0$  as  $n_0 = Z a_0$ .

The random variables  $N$  and  $A_C$  are transformed into new random variables  $\Xi$  and  $H$  and deterministic variables  $\phi(t)$  and  $\psi(t)$  such that

$$N = (\Omega Z)\phi(t) + (\Omega Z)^{1/2}\Xi \quad (10)$$

and

$$A_C = \Omega\psi(t) + \Omega^{1/2}H \quad (11)$$

where



$$\Omega = a_0$$

Consequently  $n$  and  $a_c$  are expressed as

$$n = \Omega Z \phi(t) + (\Omega Z)^{1/2} \xi \quad (12)$$

and

$$a_c = \Omega \psi(t) + \Omega^{1/2} \eta \quad (13)$$

It can be seen that

$$\frac{\partial N}{\partial \Xi} = (\Omega Z)^{1/2} \quad (14)$$

and

$$\frac{\partial A_c}{\partial H} = \Omega^{1/2} \quad (15)$$

The joint probability distribution of  $N$  and  $A_c$  is now transformed to that of  $\Xi$  and  $H$  expressed as  $\Pi(n, a_c; t)$  also denoted simply as  $\Pi$ . These are related by

$$\begin{aligned} \frac{\partial \Pi(\xi, \eta; t)}{\partial t} &= \frac{\partial P(n, a_c; t)}{\partial t} + (\Omega Z)^{1/2} \frac{d\phi}{dt} \frac{\partial \Pi(\xi, \eta; t)}{\partial \xi} \\ &\quad + \Omega^{1/2} \frac{d\psi}{dt} \frac{\partial \Pi(\xi, \eta; t)}{\partial \eta} \end{aligned} \quad (16)$$

The time derivatives of the quantities  $\xi$  and  $\eta$  are taken with constant  $n$  and  $a_c$ , i.e., in the direction given by (see, e.g., van Kampen, 1981)

$$\frac{\partial \xi}{\partial t} = -(\Omega Z)^{1/2} \frac{d\phi(t)}{dt} \quad (17)$$

and

$$\frac{\partial \eta}{\partial t} = -\Omega^{1/2} \frac{d\psi(t)}{dt} \quad (18)$$

The step operator  $E_n$  changes  $n$  into  $n+1$ , and  $E_{a_c}$ ,  $a_c$  into  $a_c+1$ ; consequently, these two operators transform  $\xi$  and  $\eta$  into  $\xi + (\Omega Z)^{-1/2}$  and  $\eta + \Omega^{1/2}$ , respectively. These operators can be expressed as (see APPENDIX A)

$$E_n = 1 + (\Omega Z)^{-1/2} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \quad (19)$$

$$E_{a_c} = 1 + \Omega^{1/2} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} + \dots \quad (20)$$

$$E_{a_c}^{-1} = 1 - \Omega^{1/2} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} - \dots \quad (21)$$

These expressions for the step operators are substituted in the master equation, equation (9). Further,  $n$  and  $a_c$  are replaced by expressions from equations (12) and (13) respectively. The terms containing  $P(n, a_c; t)$  are transformed utilizing equation (16). The master equation then assumes the following form;

$$\begin{aligned} \frac{\partial \Pi}{\partial t} - (\Omega Z)^{1/2} \frac{d\phi}{dt} \frac{\partial \Pi}{\partial \xi} - \Omega^{1/2} \frac{d\psi}{dt} \frac{\partial \Pi}{\partial \eta} \\ = \frac{\kappa}{\Omega(\Omega Z)(\Theta + \phi)} \left( (\Omega Z)^{-1/2} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \right) \\ \cdot \left( (\Omega Z)\phi + (\Omega Z)^{1/2} \xi \right) \left( \Omega - \Omega\psi - \Omega^{1/2} \eta \right) \Pi \\ + \frac{\mu}{\Omega} \left( \Omega^{1/2} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} + \dots \right) \left( \Omega\psi + \Omega^{1/2} \eta \right) \Pi \end{aligned}$$

$$\begin{aligned}
& + \frac{\lambda}{\Omega(\Omega Z)(\theta + \phi)} \left\{ \left( 1 - \Omega^{-\frac{1}{2}} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} + \dots \right) \right. \\
& \cdot \left( 1 + (\Omega Z)^{-\frac{1}{2}} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \right) - 1 \left. \right\} \\
& \cdot \left( (\Omega Z)\phi + (\Omega Z)^{\frac{1}{2}} \xi \right) \left( \Omega - \Omega \psi - \Omega^{\frac{1}{2}} \eta \right) \Pi
\end{aligned} \tag{22}$$

In this expression,  $\theta$  is the ratio of the number of oversize particles to the initial number of near mesh size particles. The quantity  $(\Omega Z)^{-\frac{1}{2}} \xi$  is negligible compared to  $\phi$ , and hence omitted in deriving the denominator of the first term on the right-hand side. The right-hand side of equation (22) is expanded by performing the multiplication throughout. To simplify further, the equation is multiplied throughout by  $\Omega$ , and the variable  $t$  is transformed into  $\tau$  defined as  $\tau = t/\Omega$ . The terms containing  $\Omega^{\frac{1}{2}}$  and  $\Omega^0$  are then collected to yield the required macroscopic equations and equations for the fluctuating components  $\xi$  and  $\eta$ , respectively (see APPENDIX B). The resultant non-linear macroscopic equation

$$\frac{d\phi}{d\tau} = - \frac{(\kappa + \lambda)\phi(1 - \psi)}{\Omega Z(\theta + \phi)} \tag{23}$$

describes the decrease in the near mesh size particle population on the sieve. The corresponding macroscopic equation of sieve blinding is

$$\frac{d\psi}{d\tau} = \frac{\lambda\phi(1 - \psi)}{\Omega(\theta + \phi)} - \frac{\mu\psi}{\Omega} \tag{24}$$

The initial conditions for these two macroscopic equations are determined by the choice of  $\phi$  and  $\psi$ . Specifically, by assuming that a fixed number of near mesh size particles and open apertures exist initially, the required conditions can be stated as  $\phi = \psi = 1$  at  $t = 0$ .

A bivariate linear Fokker-Planck equation governs the probability distribution of the fluctuating components  $\Xi$  and  $H$ , namely  $\Pi(\xi, \eta; t)$ , expressed as

$$\begin{aligned} \frac{\partial \Pi}{\partial t} = & \frac{\partial \Pi \xi}{\partial \xi} \left( \frac{(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \right) + \frac{\partial \Pi \eta}{\partial \eta} \left( \mu + \frac{\lambda \phi}{\Omega(\theta + \phi)} \right) \\ & - \frac{\partial \Pi \eta}{\partial \xi} \left( \frac{(\kappa + \lambda)\phi}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \right) - \frac{\partial \Pi \xi}{\partial \eta} \left( \frac{\lambda(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \right) \\ & + \frac{1}{2} \frac{\partial^2 \Pi}{\partial \xi^2} \left( \frac{\phi(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \right) + \frac{1}{2} \frac{\partial^2 \Pi}{\partial \eta^2} \left( \frac{\mu \psi}{\Omega} + \frac{\lambda \phi(1 - \psi)}{\Omega(\theta + \phi)} \right) \\ & - \frac{\partial^2 \Pi}{\partial \eta \partial \xi} \left( \frac{\lambda \phi(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \right) \end{aligned} \quad (25)$$

Our primary interest lies in obtaining the expressions for the moments of  $\Xi$  and  $H$  rather than an explicit solution for  $\Pi(\xi, \eta; t)$ , a bivariate, normal distribution. Hence, through further manipulation, the equations for the statistical parameters describing the joint probability distribution,  $\Pi(\xi, \eta; t)$ , are obtained as (see APPENDIX C)

$$\frac{d\langle \Xi \rangle}{dt} = - \frac{(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \langle \Xi \rangle + \frac{(\kappa + \lambda)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle H \rangle \quad (26)$$

$$\frac{d\langle H \rangle}{dt} = \frac{\lambda(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \Xi \rangle - \left( \frac{\mu}{\Omega} + \frac{\lambda \phi}{\Omega(\theta + \phi)} \right) \langle H \rangle \quad (27)$$

$$\begin{aligned} \frac{d\langle \Xi H \rangle}{dt} = & \frac{\lambda(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \Xi^2 \rangle + \frac{(\kappa + \lambda)\phi}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle H^2 \rangle \\ & - \left( \frac{(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} + \frac{\mu}{\Omega} + \frac{\lambda \phi}{\Omega(\theta + \phi)} \right) \langle \Xi H \rangle - \frac{\lambda \phi(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \end{aligned} \quad (28)$$

$$\frac{d\langle \Xi^2 \rangle}{dt} = \frac{2(\kappa + \lambda)\phi}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \Xi H \rangle - \frac{2(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \langle \Xi^2 \rangle + \frac{(\kappa + \lambda)\phi(1 - \psi)}{\Omega Z(\theta + \phi)} \quad (29)$$

$$\frac{d\langle\langle H^2 \rangle\rangle}{dt} = \frac{2\lambda(1-\psi)}{\Omega Z^{\frac{1}{2}}(\theta+\phi)} \langle\langle \Xi H \rangle\rangle - 2\left(\frac{\mu}{\Omega} + \frac{\lambda\phi(1-\psi)}{\Omega(\theta+\phi)}\right) \langle\langle H^2 \rangle\rangle$$

(30)

The initial conditions for these equations at  $t = 0$  are

$$\langle\Xi\rangle = \langle H\rangle = \langle\langle \Xi H \rangle\rangle = \langle\langle \Xi^2 \rangle\rangle = \langle\langle H^2 \rangle\rangle = 0 \quad (31)$$

where  $\langle\cdot\rangle$  represents the mean of the random variable  $(\cdot)$  and  $\langle\langle\cdot^2\rangle\rangle$  the variance;  $\langle\langle\cdot*\rangle\rangle$  is the covariance of the random variables  $(\cdot)$  and  $(*)$ . The initial conditions indicated by equation (31) are deduced from the fact (or, in some cases, an assumption) that a fixed number of particles are known to exist initially. By the choice of  $\phi$  and  $\psi$ , the means of the fluctuating components are zero, i.e.,

$$\langle\Xi\rangle = \langle H\rangle = 0$$

#### Absence of Oversize Material

The modeling of the passage of particles in the absence of oversize particles provides points of interest or reference in the sense that the master equation formulated for this case can be solved without resorting to an approximation procedure. The results obtained approximately through the system size expansion should be comparable with those obtained by solving the master equation directly.

The particles are assumed to reach the apertures at a Poisson rate. Clogging is assumed to occur as detailed previously. The transition intensity of passage of near mesh particles through the sieve is then

independent of the number of near mesh size particles but still dependent on the number of clogged apertures. The master equation can be formulated in a similar fashion as equation (9) and assumes the form

$$\begin{aligned} \frac{dP(n, a_c; t)}{dt} = & (E_n - 1) \frac{\kappa(a_o - a_c)}{a_o} P(n, a_c; t) \\ & + (E_{a_c} - 1) \frac{\mu a_c}{a_o} P(n, a_c; t) \\ & + (E_{a_c}^{-1} E_n - 1) \frac{\lambda(a_o - a_c)}{a_o} P(n, a_c; t) \end{aligned} \quad (32)$$

The governing differential equations for the moments of the random variables  $N$  and  $A_c$  are derived from this expression. The equations for the means and variances of  $N$  and  $A_c$  and their covariance are obtained by multiplying both sides of the master equation by the respective parameters, summing over all possible values, and utilizing the property of the step operator,

$$\sum_n \sum_{a_c} g(n, a_c) E_n^{-1} E_{a_c}^{-1} f(n, a_c) = \sum_n \sum_{a_c} f(n, a_c) E_n^{-1} E_{a_c}^{-1} g(n, a_c) \quad (33)$$

For instance, an equation for the average value of  $N$  is obtained by multiplying throughout by  $n$  and summing over all possible values of  $n$  and  $a_c$ . This procedure yields the following expressions (see APPENDIX D):

$$\frac{d\langle N \rangle}{dt} = - \frac{(\kappa + \lambda)(a_o - \langle A_c \rangle)}{a_o} \quad (34)$$

$$\frac{d\langle A_c \rangle}{dt} = \frac{\lambda(a_o - \langle A_c \rangle)}{a_o} - \frac{\mu \langle A_c \rangle}{a_o} \quad (35)$$

$$\frac{d\langle\langle N_c^2 \rangle\rangle}{dt} = \frac{2(\kappa+\lambda)\langle\langle NA_c \rangle\rangle}{a_o} + (\kappa+\lambda)\left(1 - \frac{\langle A_c \rangle}{a_o}\right) \quad (36)$$

$$\frac{d\langle\langle A_c^2 \rangle\rangle}{dt} = -\frac{2(\lambda+\mu)\langle\langle A_c^2 \rangle\rangle}{a_o} + \lambda\left(1 - \frac{\langle A_c \rangle}{a_o}\right) + \frac{\mu\langle A_c \rangle}{a_o} \quad (37)$$

$$\frac{d\langle\langle NA_c \rangle\rangle}{dt} = \frac{(\kappa+\lambda)\langle\langle A_c^2 \rangle\rangle}{a_o} - \frac{(\lambda+\mu)\langle\langle NA_c \rangle\rangle}{a_o} - \lambda\left(1 - \frac{\langle A_c \rangle}{a_o}\right) \quad (38)$$

with the initial conditions

$$\langle N \rangle = n_o, \quad \langle A_c \rangle = \langle\langle N \rangle\rangle = \langle\langle A_c \rangle\rangle = \langle\langle NA_c \rangle\rangle = 0 \text{ at } t = 0 \quad (39)$$

Equations (34) and (35) are solved to yield the following analytical expressions for the mean number of near mesh size particles on the sieve and mean number of clogged apertures, respectively.

$$\langle N \rangle = n_o - \frac{(\kappa+\lambda)\mu t}{\lambda+\mu} - \frac{(\kappa+\lambda)\lambda a_o}{(\lambda+\mu)^2} \left[ 1 - \exp\left\{-\frac{(\lambda+\mu)t}{a_o}\right\} \right] \quad (40)$$

$$\langle A_c \rangle = \frac{\lambda a_o}{(\lambda+\mu)} \left[ 1 - \exp\left\{-\frac{(\lambda+\mu)t}{a_o}\right\} \right] \quad (41)$$

#### NUMERICAL SIMULATION

The non-linear macroscopic equations, equations (23) and (24), and the equations for the moments of the probability distribution  $\Pi(\xi, \eta; t)$ , equations (28) through (30), do not yield explicit analytical solutions. A fourth order Runge Kutta algorithm, therefore, has been employed to solve these equations simultaneously to simulate the sieving operation numerically. The variation in the behavior of the system with respect to the number of particles passing through the sieve and the number of

apertures blinded has been investigated for various values of the sieving rate parameters, i.e., the transition rate constants  $\kappa$ ,  $\lambda$  and  $\mu$ . This is repeated for different values of the system parameters  $a_0$ ,  $n_0$ , and  $n_{os}$ . The rate of decrease in the near mesh size particles present on the sieve, represented by the macroscopic equation (23) is also studied for special cases, e.g., the case with a high initial proportion of near mesh size particles ( $n_0 \gg n_{os}$ ).

Another situation analyzed is that representing the terminal stages of sieving, i.e., passage of near mesh size particles in the presence of a comparatively large number of oversize particles ( $n_0 \ll n_{os}$ ). The equations derived so far are assumed to be valid until the number of active particles present on the sieve surface becomes less than or equal to the number of open apertures.

#### RESULTS AND DISCUSSION

The results of the numerical simulation resorting to the system size expansion, and those by the direct solutions for the case without oversize particles are plotted in Figures 1 to 12. The covariance,  $\langle\langle\xi\eta\rangle\rangle$ , has been plotted along with the correlation coefficient  $\rho$ , where  $\rho$  is defined as

$$\rho = \frac{\langle\langle\xi\eta\rangle\rangle}{\sqrt{\langle\langle\xi^2\rangle\rangle\langle\langle\eta^2\rangle\rangle}} \quad (42)$$

for the solution of the master equation using the system size expansion in Figures 4 and 8, and as



$$\rho = \frac{\langle\langle NA \rangle\rangle}{\sqrt{\langle\langle N^2 \rangle\rangle \langle\langle A_c^2 \rangle\rangle}} \quad (43)$$

for the case without oversize particles in Figure 12.

The mean and variance of the number of particles passing through the sieve denoted by  $\langle X \rangle$  and  $\langle\langle X^2 \rangle\rangle$ , respectively, are related to the mean and variance of  $N$  and  $A_c$  in the following manner

$$\langle X \rangle = n_o - \phi n_o - \psi a_o \quad (44)$$

$$\langle\langle X^2 \rangle\rangle = \langle\langle \Xi^2 \rangle\rangle n_o + \langle\langle H^2 \rangle\rangle a_o \quad (45)$$

for the case of the system size expansion and

$$\langle X \rangle = 1 - \langle N \rangle - \langle A_c \rangle \quad (46)$$

$$\langle\langle X^2 \rangle\rangle = \langle\langle N^2 \rangle\rangle + \langle\langle A_c^2 \rangle\rangle \quad (47)$$

for the case without oversize particles. These quantities are plotted as fractions of the initial near mesh size particle population along with the other variables as functions of time.

The application of the present model requires the knowledge of the number of oversize particles, sieve openings, and the initial number of near mesh size particles. Data on sieving operations in the literature lacks this information. However, the results of many studies are predicted, at least qualitatively, by this model.

The passage of particles through a 48 mesh sieve in the presence of clogging and oversize material has been studied by Kapur et al. (1977), using a Rotap device and sonic sifter. The model and the corresponding numerical simulation deal with the terminal stages of sieving. The

exponential decay of the near mesh size particle population presented in their results is predicted by the present model. This situation is described in Figures 1 and 2. The authors have assumed the choking phenomenon to be a poisson process in presenting a simplified stochastic model. However, the transition intensities concerning the choking phenomenon are derived here from probabilistic considerations of particle behavior. The description of the variation of the number of clogged apertures with time is presented in Figure 3. It matches qualitatively with that of Standish (1985) up to a point in time after which the number of particles on the sieve are equal to or less than the number of open apertures. This indicates the validity of the transition intensities. The situation with a high proportion of near mesh size particles is described in Figures 5 through 8. The passage of particles in the absence of oversize material is described in Figures 9 through 12.

Standish (1985) has pointed out that oversize material actually could be beneficial to the passage of near mesh size particles, contrary to previous observations and the present model. Nevertheless, as can be inferred from the description and photographs presented in his paper, this situation occurs when very few particles are left on the sieve surface; however, it is not significant in practice. The 'nudging effect' of the oversize material at this stage of the sieving operation cannot be considered to prevail during the major portion of the sieving operation.

CONCLUDING REMARKS

Several significant observations can be made based on the results obtained. The presence of oversize particles decreases the sieving rate of near mesh size particles. For a population with a high proportion of near mesh size particles, the flow rate is almost constant. During the terminal stages of sieving, exponential decay is observed for the particle population retained on the sieve. When the particle population reaches a size equal to or below that of the open apertures, different conditions exist; they are characterized by a stochastically independent behavior of the particles with a dependence only on the number of particles retained on the sieve for both passage and sieve blinding. The number of particles retained then may follow an exponential decay.

An examination of the fluctuations indicates that as expected, the fluctuations are larger for a smaller particle population. In the presence of oversize particles, the distribution of the number of particles evolves into approximately a Poisson distribution when the flow rate is dependent on the number of particles. With no explicit dependence of the flow rate on the number of particles present, as in the case without oversize particles, the variance increases beyond the mean value with time. This is expected as small deviations in the number of clogged apertures give rise to large fluctuations in the distribution of the number of particles remaining on the sieve. The description of the fluctuations provides means to determine an optimal sieving time and information for improved sieving specifications. The present stochastic model is thus a valuable tool for describing the inherent random processes that prevail during the sieving operation. Future research in

sieving would prove beneficial only if the refined methodology of particle size classification and inclusion of the inherent fluctuations is followed as indicated in the present study.

# NOTATION

$A_c, a_c'$  = random variable representing the number of blinded apertures

$a_c$  = value assumed by random variable  $A_c$

$N$  = random variable representing the number of near mesh size particles on the sieve

$n, n'$  = values assumed by  $N$

$n_o$  = initial number of near mesh size particles

$n_{os}$  = number of oversize particles

$P(n, a_c; t)$  = joint probability distribution of  $N$  and  $A_c$

## Greek Letters

$Z$  =  $n_o/a_o$

$H$  = fluctuating component of  $A_c$

$\eta$  = value assumed by  $H$

$\theta$  =  $n_{os}/n_o$

$\kappa_{n, a_c}$  = intensity of transition of passage of a near mesh size particle through the sieve,  $\text{time}^{-1}$

$\kappa$  = proportionality constant in the intensity of transition,  $\kappa_{n, a_c}$ ,  $\text{time}^{-1}$

$\lambda_{n, a_c}$  = intensity of transition of blinding of a sieve opening,  $\text{time}^{-1}$

$\lambda$  = proportionality constant in the intensity of transition  $\lambda_{n, a_c}$ ,  $\text{time}^{-1}$

$\mu_{a_c}$	= intensity of transition of a trapped particle passing through a sieve opening, $\text{time}^{-1}$
$\mu$	= proportionality constant in the intensity of transition $\mu_{a_c}$ , $\text{time}^{-1}$
$\Xi$	= fluctuating component of N
$\xi$	= value assumed by $\Xi$
$\Pi(\xi, \eta; t)$	= probability density function of $\xi$ and $\eta$
$\rho$	= correlation coefficient
$\phi$	= macroscopic component of N
$\psi$	= macroscopic component of $A_c$
$\Omega$	= system size parameter
$\langle \cdot \rangle$	= expected value or mean of random variable
$\langle \langle \cdot^2 \rangle \rangle$	= variance of random variable
$\langle \langle \cdot * \rangle \rangle$	= covariance between two random variables
$E$	= step operator

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# APPENDIX A. EXPANSION OF THE STEP OPERATOR

The probability distribution  $P(n, a_c; t)$  is transformed into  $\Pi(\xi, \eta; t)$  by expressing  $n$  and  $a_c$  as

$$n = n_0 \phi + n_0^{-1/2} \xi \quad (A1)$$

$$a_c = a_0 \psi + a_0^{-1/2} \eta \quad (A2)$$

respectively. This yields

$$P(n, a_c; t) \Delta n = \Pi(\xi, \eta; t) \Delta \xi \quad (A3)$$

$$P(n, a_c; t) \Delta a_c = \Pi(\xi, \eta; t) \Delta \eta \quad (A4)$$

For instance, a unit increase in  $n$  can be seen to be equivalent to the increase in  $\xi$  of  $n_0^{-1/2}$ . Expanding  $\Pi(\xi + n_0^{-1/2}, \eta; t)$  with respect to  $\xi$  in a Taylor's series yields

$$\begin{aligned} \Pi(\xi + n_0^{-1/2}, \eta; t) &= \Pi(\xi, \eta; t) + n_0^{-1/2} \frac{\partial}{\partial \xi} \Pi(\xi, \eta; t) \\ &\quad + \frac{1}{2} n_0^{-1} \frac{\partial^2}{\partial \xi^2} \Pi(\xi, \eta; t) + \dots \\ &= (1 + n_0^{-1/2} \frac{\partial}{\partial \xi} + \frac{1}{2} n_0^{-1} \frac{\partial^2}{\partial \xi^2} + \dots) \Pi(\xi, \eta; t), \end{aligned} \quad (A5)$$

i.e.,

$$P(n+1, a_c; t) = (1 + n_0^{-1/2} \frac{\partial}{\partial \xi} + \frac{1}{2} n_0^{-1} \frac{\partial^2}{\partial \xi^2} + \dots) P(n, a_c; t) \quad (A6)$$

Since

$$E_n P(n, a_c; t) = P(n+1, a_c; t),$$

we have



$$\mathbb{E}_n = 1 + n_0^{-1/2} \frac{\partial}{\partial \xi} + \frac{1}{2} n_0^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \quad (\text{A7})$$

Similarly, we obtain the following:

$$\mathbb{E}_{a_c} = 1 + a_0^{-1/2} \frac{\partial}{\partial \eta} + \frac{1}{2} a_0^{-1} \frac{\partial^2}{\partial \eta^2} + \dots \quad (\text{A8})$$

$$\mathbb{E}_{a_c}^{-1} = 1 - a_0^{-1/2} \frac{\partial}{\partial \eta} + \frac{1}{2} a_0^{-1} \frac{\partial^2}{\partial \eta^2} + \dots \quad (\text{A9})$$

# APPENDIX B. EXPANSION OF EQUATION (22) IN THE TEXT

Equation (22) in the text is multiplied throughout by the system size parameter  $\Omega$  with the variable  $t$  transformed into  $\tau$  defined as  $\tau=t/\Omega$ . The resultant equation assumes the form

$$\begin{aligned} & \frac{\partial \Pi}{\partial \tau} - (\Omega Z)^{\frac{1}{2}} \frac{d\phi}{d\tau} \frac{\partial \Pi}{\partial \xi} - \Omega^{\frac{1}{2}} \frac{\partial \psi}{\partial \tau} \frac{\partial \Pi}{\partial \eta} \\ &= \frac{\kappa}{Z(\theta+\phi)} \left( (\Omega Z)^{-\frac{1}{2}} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \right) (\Omega Z \phi + (\Omega Z)^{\frac{1}{2}} \xi) \\ & \quad \cdot (1 - \psi - \Omega^{-\frac{1}{2}} \eta) \Pi \\ & \quad + \mu \left( \Omega^{-\frac{1}{2}} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} \right) (\Omega \psi + \Omega^{\frac{1}{2}} \eta) \Pi \\ & \quad + \frac{\lambda}{Z(\theta+\phi)} \left\{ (1 + (\Omega Z)^{-\frac{1}{2}} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots) \right. \\ & \quad \cdot (1 - \Omega^{-\frac{1}{2}} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} + \dots)^{-1} \\ & \quad \cdot (\Omega Z \phi + (\Omega Z)^{\frac{1}{2}} \xi) (1 - \psi - \Omega^{-\frac{1}{2}} \eta) \Pi \end{aligned} \quad (B1)$$

Expansion of the right-hand side by multiplying the corresponding quantities yields

$$\begin{aligned} & \frac{\partial \Pi}{\partial \tau} - (\Omega Z)^{\frac{1}{2}} \frac{d\phi}{d\tau} - \Omega^{\frac{1}{2}} \frac{\partial \psi}{\partial \tau} \frac{\partial \Pi}{\partial \eta} \\ &= \frac{\kappa}{Z(\theta+\phi)} \left( (\Omega Z)^{-\frac{1}{2}} \frac{\partial}{\partial \xi} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} + \dots \right) \\ & \quad \cdot ( (\Omega Z) \phi (1-\psi) - \Omega^{\frac{1}{2}} Z \phi \eta + (\Omega Z)^{\frac{1}{2}} (1-\psi) \xi - Z^{\frac{1}{2}} \xi \eta ) \Pi \\ & \quad + \mu \left( \Omega^{\frac{1}{2}} \psi \frac{\partial \Pi}{\partial \eta} + \frac{\partial \Pi \eta}{\partial \eta} + \frac{1}{2} \psi \frac{\partial^2 \Pi}{\partial \eta^2} + \frac{1}{2} \Omega^{-\frac{1}{2}} \frac{\partial^2 \eta \Pi}{\partial \eta^2} + \dots \right) \end{aligned}$$

$$\begin{aligned}
& + \frac{\lambda}{Z(\Theta+\Phi)} \left( -\Omega^{-\frac{1}{2}} \frac{\partial}{\partial \eta} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \eta^2} + (\Omega Z)^{-\frac{1}{2}} \frac{\partial}{\partial \xi} - \Omega^{-1} Z^{-\frac{1}{2}} \frac{\partial^2}{\partial \eta \partial \xi} \right. \\
& + \frac{1}{2} \Omega^{-3/2} Z^{-\frac{1}{2}} \frac{\partial^3}{\partial \xi \partial \eta^2} + \frac{1}{2} (\Omega Z)^{-1} \frac{\partial^2}{\partial \xi^2} - \frac{1}{2} \Omega^{-3/2} Z^{-1} \frac{\partial^3}{\partial \xi^2 \partial \eta} \\
& \left. + \frac{1}{4} \Omega^{-2} Z^{-1} \frac{\partial^4}{\partial \xi^2 \partial \eta^2} + \dots \right) \\
& \cdot \left( (\Omega Z \Phi (1-\psi) - \Omega^{\frac{1}{2}} Z \Phi \eta + (\Omega Z)^{\frac{1}{2}} (1-\psi) \xi - Z^{\frac{1}{2}} \xi \eta) \Pi \right) \quad (B2)
\end{aligned}$$

Further manipulation yields

$$\begin{aligned}
& \frac{\partial \Pi}{\partial \tau} - (\Omega Z)^{\frac{1}{2}} \frac{d\Phi}{d\tau} - \Omega^{\frac{1}{2}} \frac{\partial \psi}{\partial \tau} \frac{\partial \Pi}{\partial \eta} \\
= & \frac{\kappa}{Z(\Theta+\Phi)} \left( (\Omega Z)^{\frac{1}{2}} \Phi (1-\psi) \frac{\partial \Pi}{\partial \xi} - Z^{\frac{1}{2}} \Phi \frac{\partial \Pi \eta}{\partial \eta} + (1-\psi) \frac{\partial \Pi \xi}{\partial \xi} - \Omega^{-\frac{1}{2}} \frac{\partial \Pi \xi \eta}{\partial \xi} \right. \\
& + \frac{1}{2} \Phi (1-\psi) \frac{\partial^2 \Pi}{\partial \xi^2} - \frac{1}{2} \Omega^{-\frac{1}{2}} \Phi \frac{\partial^2 \Pi \eta}{\partial \xi^2} + \frac{1}{2} (\Omega Z)^{-\frac{1}{2}} (1-\psi) \frac{\partial^2 \Pi \xi}{\partial \xi^2} \\
& \left. - \frac{1}{2} \Omega^{-1} Z^{-\frac{1}{2}} \frac{\partial^2 \Pi \xi \eta}{\partial \xi^2} + \dots \right) \\
& + \mu \left( \Omega^{\frac{1}{2}} \psi \frac{\partial \Pi}{\partial \eta} + \frac{\partial \Pi \eta}{\partial \eta} + \frac{1}{2} \psi \frac{\partial^2 \Pi}{\partial \eta^2} + \frac{1}{2} \Omega^{-\frac{1}{2}} \frac{\partial^2 \Pi \eta}{\partial \eta^2} + \dots \right) \\
& + \frac{\lambda}{Z(\Theta+\Phi)} \left( -\Omega^{\frac{1}{2}} Z \Phi (1-\psi) \frac{\partial \Pi}{\partial \eta} + Z \Phi \frac{\partial \Pi \eta}{\partial \eta} - Z^{\frac{1}{2}} (1-\psi) \frac{\partial \xi \Pi}{\partial \eta} + \Omega^{-\frac{1}{2}} Z^{\frac{1}{2}} \frac{\partial \xi \eta \Pi}{\partial \eta} \right. \\
& + \frac{1}{2} Z \Phi (1-\psi) \frac{\partial^2 \Pi}{\partial \eta^2} - \frac{1}{2} \Omega^{-\frac{1}{2}} Z \Phi \frac{\partial^2 \Pi \eta}{\partial \eta^2} \\
& + \frac{1}{2} \Omega^{-\frac{1}{2}} Z^{\frac{1}{2}} (1-\psi) \frac{\partial^2 \Pi \xi}{\partial \eta^2} - \frac{1}{2} \Omega^{-1} Z^{\frac{1}{2}} \frac{\partial^2 \xi \eta \Pi}{\partial \eta^2} \\
& + (\Omega Z)^{\frac{1}{2}} \Phi (1-\psi) \frac{\partial \Pi}{\partial \xi} - Z^{\frac{1}{2}} \Phi \frac{\partial \Pi \eta}{\partial \xi} + (1-\psi) \frac{\partial \Pi \xi}{\partial \xi} - \Omega^{-\frac{1}{2}} \frac{\partial \xi \eta \Pi}{\partial \xi} \\
& \left. - Z^{\frac{1}{2}} \Phi (1-\psi) \frac{\partial^2 \Pi}{\partial \eta \partial \xi} + \Omega^{-\frac{1}{2}} Z^{\frac{1}{2}} \Phi \frac{\partial^2 \Pi \eta}{\partial \eta \partial \xi} - \Omega^{-\frac{1}{2}} (1-\psi) \frac{\partial^2 \Pi \xi}{\partial \eta \partial \xi} \right)
\end{aligned}$$

$$\begin{aligned}
& - \Omega^{-1} \frac{\partial^2 \xi \eta \Pi}{\partial \eta \partial \xi} + \frac{1}{2} \Omega^{-\frac{1}{2}} Z^{\frac{1}{2}} \phi(1-\psi) \frac{\partial^2 \Pi}{\partial \xi \partial \eta^2} - \frac{1}{2} \Omega^{-1} Z^{\frac{1}{2}} \phi \frac{\partial^3 \eta \Pi}{\partial \xi \partial \eta^2} \\
& + \frac{1}{2} \Omega^{-\frac{1}{2}} (1-\psi) \frac{\partial^3 \Pi \xi}{\partial \xi \partial \eta^2} - \frac{1}{2} \Omega^{-3/2} \frac{\partial^3 \xi \eta \Pi}{\partial \xi \partial \eta^2} + \frac{1}{2} \phi(1-\psi) \frac{\partial^2 \Pi}{\partial \xi^2} \\
& - \frac{1}{2} \Omega^{-\frac{1}{2}} \phi \frac{\partial^2 \Pi \eta}{\partial \xi^2} + \frac{1}{2} (\Omega Z)^{-\frac{1}{2}} (1-\psi) \frac{\partial^2 \Pi \xi}{\partial \xi^2} - \frac{1}{2} \Omega^{-1} Z^{-\frac{1}{2}} \frac{\partial^2 \xi \eta \Pi}{\partial \xi^2} \\
& - \frac{1}{2} \Omega^{-\frac{1}{2}} \phi(1-\psi) \frac{\partial^3 \Pi}{\partial \xi^2 \partial \eta} + \frac{1}{2} \Omega^{-1} \phi \frac{\partial^2 \eta \Pi}{\partial \xi^2 \partial \eta} - \frac{1}{2} \Omega^{-1} Z^{-\frac{1}{2}} (1-\psi) \\
& \frac{\partial^3 \Pi \xi}{\partial \xi^2 \partial \eta} \\
& + \frac{1}{2} \Omega^{-3/2} Z^{-\frac{1}{2}} \frac{\partial^3 \xi \eta \Pi}{\partial \xi^2 \partial \eta} + \frac{1}{4} \Omega^{-1} \phi(1-\psi) \frac{\partial^4 \Pi}{\partial \xi^2 \partial \eta^2} - \frac{1}{4} \Omega^{-3/2} \phi \\
& \frac{\partial^4 \Pi \eta}{\partial \xi^2 \partial \eta^2} \\
& + \frac{1}{4} \Omega^{-3/2} Z^{-\frac{1}{2}} (1-\psi) \frac{\partial^4 \Pi \xi}{\partial \xi^2 \partial \eta^2} - \frac{1}{4} \Omega^{-2} Z^{-\frac{1}{2}} \frac{\partial^4 \xi \eta \Pi}{\partial \xi^2 \partial \eta^2} + \dots )
\end{aligned}$$

(B3)

Now, the terms on the left-hand side and those on the right-hand side containing  $\Omega^{\frac{1}{2}}$  are collected; equating these terms results in

$$\begin{aligned}
& -(\Omega Z)^{\frac{1}{2}} \frac{d\phi}{d\tau} \frac{\partial \Pi}{\partial \xi} - \Omega^{\frac{1}{2}} \frac{d\psi}{d\tau} \frac{\partial \Pi}{\partial \eta} \\
& = (\Omega Z)^{\frac{1}{2}} \frac{\kappa \phi(1-\psi)}{Z(\theta+\phi)} \frac{\partial \Pi}{\partial \xi} + (\Omega Z)^{\frac{1}{2}} \frac{\lambda \phi(1-\psi)}{Z(\theta+\phi)} \frac{\partial \Pi}{\partial \xi} + \Omega^{\frac{1}{2}} \mu \psi \frac{\partial \Pi}{\partial \eta} - \Omega^{\frac{1}{2}} \frac{\lambda Z \phi(1-\psi)}{Z(\theta+\phi)} \frac{\partial \Pi}{\partial \eta}
\end{aligned}$$

(B4)

Note that all terms in this expression are either proportional to  $\partial \Pi / \partial \xi$  or to  $\partial \Pi / \partial \eta$ . The coefficient of the former vanishes if

$$\frac{d\phi}{dt} = - \frac{(\kappa+\lambda)\phi(1-\psi)}{\Omega Z(\theta+\phi)} \quad (B5)$$

and that of the latter vanishes if

$$\frac{d\psi}{dt} = - \frac{\mu\psi}{\Omega} + \frac{\lambda\phi(1-\psi)}{\Omega(\theta+\phi)} \quad (B6)$$

which are the macroscopic equations, equations (23) and (24), respectively, in the text. In a similar fashion, equation (25) in the text is obtained by collecting the terms of the order of  $\Omega^0$  and regrouping.

APPENDIX C. LINEAR FOKKER-PLANCK EQUATION AND DERIVATION OF THE MOMENTS  
OF THE PROBABILITY DISTRIBUTION

The generalized linear multivariate Fokker Plank equation has the form

$$\frac{\partial P(\xi; t)}{\partial t} = - \sum_{i,j} A_{ij} \frac{\partial \xi_j P(\xi; t)}{\partial \xi_i} + \frac{1}{2} \sum_{i,j} B_{ij} \frac{\partial^2 P(\xi; t)}{\partial \xi_i \partial \xi_j} \quad (C1)$$

where  $A_{ij}$  and  $B_{ij}$  are the coefficient matrices and  $P(\xi; t)$  (or  $P$ ) is the general probability distribution of the vector  $\xi$  with components  $\xi_i$  (see, e.g., van Kampen, 1981). The linear bivariate Fokker Plank equation obtained in the text, equation (25), is written in terms of  $\xi, \eta$  and  $\Pi(\xi, \eta; t)$  (or  $\Pi$ ) as

$$\begin{aligned} \frac{\partial \Pi}{\partial t} = & - \left( A_{\xi\xi} \frac{\partial \xi \Pi}{\partial \xi} + A_{\xi\eta} \frac{\partial \eta \Pi}{\partial \xi} + A_{\eta\xi} \frac{\partial \xi \Pi}{\partial \eta} + A_{\eta\eta} \frac{\partial \eta \Pi}{\partial \eta} \right) \\ & + \frac{1}{2} \left( B_{\xi\xi} \frac{\partial^2 \Pi}{\partial \xi^2} + B_{\xi\eta} \frac{\partial^2 \Pi}{\partial \xi \partial \eta} + B_{\eta\xi} \frac{\partial^2 \Pi}{\partial \eta \partial \xi} + B_{\eta\eta} \frac{\partial^2 \Pi}{\partial \eta^2} \right) \end{aligned} \quad (C2)$$

Note that the coefficients are time dependent. The expressions for the first and second moments are derived as follows:

Let  $f(\xi, \eta)$  and  $g(\xi, \eta)$  be two functions of  $\xi$  and  $\eta$ . The sum of  $g(\xi, \eta) \frac{\partial}{\partial \xi} f(\xi, \eta)$  over all possible values of  $\xi$  can be expressed in terms of the central difference approximation as

$$\begin{aligned} & \sum g(\xi, \eta) \frac{\partial}{\partial \xi} f(\xi, \eta) \\ & \approx \sum g(\xi, \eta) \left[ \frac{f(\xi + \Delta\xi, \eta) - f(\xi, \eta)}{\Delta\xi} \right] \\ & = \frac{1}{\Delta\xi} [\sum g(\xi, \eta) f(\xi + \Delta\xi, \eta) - \sum g(\xi, \eta) f(\xi, \eta)] \end{aligned} \quad (C3)$$

The quantity  $\Sigma g(\xi, \eta) f(\xi + \Delta \xi, \eta)$  can be transformed utilizing the step operator properties such as (also see APPENDIX D)

$$\Sigma g(\xi, \eta) f(\xi + \Delta \xi, \eta) = \Sigma f(\xi, \eta) g(\xi - \Delta \xi, \eta) \quad (C4)$$

which, in conjunction with equation (C3), yields

$$\Sigma g(\xi, \eta) \frac{\partial}{\partial \xi} f(\xi, \eta) = - \Sigma f(\xi, \eta) \frac{\partial}{\partial \xi} g(\xi, \eta) \quad (C5)$$

A similar treatment with respect to the variable  $\eta$ , and the second order partial derivative of  $f(\xi, \eta)$  yields

$$\Sigma g(\xi, \eta) \frac{\partial}{\partial \eta} f(\xi, \eta) = - \Sigma f(\xi, \eta) \frac{\partial}{\partial \eta} g(\xi, \eta) \quad (C6)$$

$$\Sigma g(\xi, \eta) \frac{\partial^2}{\partial \xi^2} f(\xi, \eta) = \Sigma f(\xi, \eta) \frac{\partial^2}{\partial \xi^2} g(\xi, \eta) \quad (C7)$$

$$\Sigma g(\xi, \eta) \frac{\partial^2}{\partial \eta^2} f(\xi, \eta) = \Sigma f(\xi, \eta) \frac{\partial^2}{\partial \eta^2} g(\xi, \eta) \quad (C8)$$

To obtain the equations for the means and variances of  $\xi$  and  $\eta$ , and their covariance, equation (C2) is multiplied by the respective parameters and summed over all possible values, and equations (C5) through (C8) are utilized to obtain the required relationship. For illustration, the equation for  $\xi$  is derived as follows: Multiplying equation (C2) by  $\xi$  and summing the resultant expression over all values of  $\xi$  and  $\eta$ , we have

$$\begin{aligned} \frac{d\langle \xi \rangle}{dt} = & - A_{\xi \xi} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial \xi \Pi}{\partial \xi} - A_{\xi \eta} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial \eta \Pi}{\partial \xi} \\ & - A_{\eta \xi} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial \xi \Pi}{\partial \eta} - A_{\eta \eta} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial \eta \Pi}{\partial \eta} \\ & + \frac{1}{2} B_{\xi \xi} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial^2 \Pi}{\partial \xi^2} + \frac{1}{2} B_{\xi \eta} \Sigma_{\eta} \Sigma_{\xi} \xi \frac{\partial^2 \Pi}{\partial \xi \partial \eta} \end{aligned}$$

$$+ \frac{1}{2} B_{\eta\eta} \sum_{\eta} \sum_{\xi} \xi \frac{\partial^2 \pi}{\partial \eta^2} + \frac{1}{2} B_{\eta\xi} \sum_{\eta} \sum_{\xi} \xi \frac{\partial^2 \pi}{\partial \eta \partial \xi} \quad (C9)$$

Employing equations (C5) through (C8), equation (C9) transforms into

$$\frac{d\langle \xi \rangle}{dt} = - \left( A_{\xi\xi} \langle \xi \rangle + A_{\xi\eta} \langle \eta \rangle \right) \quad (C10)$$

which has the initial condition at  $t = 0$ ,  $\langle \xi \rangle = 0$ . In terms of the various quantities represented by the coefficients, equation (C10) can be rewritten as

$$\frac{d\langle \xi \rangle}{dt} = - \frac{(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \langle \xi \rangle + \frac{(\kappa + \lambda)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \eta \rangle \quad (C11)$$

A similar expression can be derived for the mean value of the fluctuating component of  $A_c$ , i.e.,

$$\frac{d\langle \eta \rangle}{dt} = \frac{\lambda(1 - \psi)}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \xi \rangle - \left( \frac{\mu}{\Omega} + \frac{\lambda\phi}{\Omega(\theta + \phi)} \right) \langle \eta \rangle \quad (C12)$$

with an initial condition of  $\langle \eta \rangle = 0$ . Through the choice of  $\phi$  and  $\psi$  and the knowledge of their fixed values at time  $t=0$ , it can be easily seen that the mean of the fluctuating components is zero, i.e.,

$$\langle \xi \rangle = \langle \eta \rangle = 0$$

To obtain the variances of  $\xi$  and  $\eta$  and their covariance, equation (C9) is multiplied by  $\xi^2$ , and the resultant expression is summed over all possible values of  $\xi$  and  $\eta$  and transformed by utilizing equations (C5) through (C8). This results in the following equation for the variance of  $\xi$ .

$$\frac{d\langle \xi^2 \rangle}{dt} = \frac{-2(\kappa + \lambda)(1 - \psi)}{\Omega Z(\theta + \phi)} \langle \xi^2 \rangle + \frac{2(\kappa + \lambda)\phi}{\Omega Z^{\frac{1}{2}}(\theta + \phi)} \langle \xi \eta \rangle + \frac{(\kappa + \lambda)\phi(1 - \psi)}{\Omega Z(\theta + \phi)}$$



(C13)

To obtain the covariance of  $\xi$  and  $\eta$ , equation (C9) is multiplied by  $\xi\eta$  and the resultant expression is summed over all possible values of  $\xi$  and  $\eta$ . By resorting to equations (C5) through (C8), we eventually obtain

$$\begin{aligned} \frac{d\langle\langle\xi\eta\rangle\rangle}{dt} = & + \frac{\lambda(1-\psi)}{\Omega Z^{\frac{1}{2}}(\theta+\phi)} \langle\langle\xi^2\rangle\rangle - \left( \frac{(\kappa+\lambda)(1-\psi)}{\Omega Z(\theta+\phi)} + \frac{\mu}{\Omega} + \frac{\lambda\phi}{\Omega(\theta+\phi)} \right) \langle\langle\xi\eta\rangle\rangle \\ & + \frac{(\kappa+\lambda)\phi}{\Omega Z^{\frac{1}{2}}(\theta+\phi)} \langle\langle\eta^2\rangle\rangle - \frac{\lambda\phi(1-\psi)}{\Omega Z^{\frac{1}{2}}(\theta+\phi)} \end{aligned} \quad (C14)$$

Finally, the governing differential equation for the variance of  $\eta$  is obtained similarly as

$$\begin{aligned} \frac{d\langle\langle\eta^2\rangle\rangle}{dt} = & \frac{2\lambda(1-\psi)}{\Omega Z^{\frac{1}{2}}(\theta+\phi)} \langle\langle\xi\eta\rangle\rangle - 2\left( \frac{\mu}{\Omega} + \frac{\lambda\phi}{\Omega(\theta+\phi)} \right) \langle\langle\eta^2\rangle\rangle \\ & + \frac{\mu\psi}{\Omega} + \frac{\lambda\phi(1-\psi)}{\Omega(\theta+\phi)} \end{aligned} \quad (C15)$$

The initial conditions for these three equations are  $\langle\langle\xi^2\rangle\rangle = \langle\langle\eta^2\rangle\rangle = \langle\langle\xi\eta\rangle\rangle = 0$  at  $t = 0$ .

# APPENDIX D. DIRECT SOLUTION TO THE MASTER EQUATION

The master equation, for the case of absence of oversize particles, as given by equation (29) in the text, is

$$\begin{aligned} \frac{dP(n, a_c; t)}{dt} = & (E_n - 1) \frac{\kappa(a_o - a_c)}{a_o} P(n, a_c; t) \\ & + (E_{a_c} - 1) \frac{\mu a_c}{a_o} P(n, a_c; t) \\ & + (E_n E_{a_c}^{-1} - 1) \frac{\lambda(a_o - a_c)}{a_o} P(n, a_c; t) \end{aligned} \quad (D1)$$

To obtain the equation for the average value of the number of near mesh size particles retained on the sieve surface, for instance, equation (D1) is multiplied by  $n$ , and each term in the resultant expression is summed over all possible values of  $n$  and  $a_c$ ; this yields

$$\begin{aligned} \sum_n \sum_{a_c} n \frac{dP(n, a_c; t)}{dt} = & \sum_n \sum_{a_c} n (E_n - 1) \kappa \frac{(a_o - a_c)}{a_o} P(n, a_c; t) \\ & + \sum_n \sum_{a_c} n (E_{a_c} - 1) \frac{\mu a_c}{a_o} P(n, a_c; t) \\ & + \sum_n \sum_{a_c} n (E_n E_{a_c}^{-1} - 1) \frac{\lambda(a_o - a_c)}{a_o} P(n, a_c; t) \end{aligned} \quad (D2)$$

The summation being over the state space variables  $n$  and  $a_c$  and not with respect to  $t$ , it can be moved within the derivative with regard to the terms on the left-hand side of the equation. The terms on the right hand side can be manipulated using the property of the step operator, specifically,

$$\sum_n \sum_{a_c} g(n, a_c) E_n^{-1} E_{a_c}^{-1} f(n, a_c) = \sum_n \sum_{a_c} f(n, a_c) E_n^{-1} E_{a_c}^{-1} g(n, a_c) \quad (D3)$$

for any pair of functions  $f(n, a_c)$  and  $g(n, a_c)$ . This results in

$$\begin{aligned} & \frac{d \sum_n \sum_{a_c} n P(n, a_c; t)}{dt} \\ = & \frac{K}{a_o} \sum_n \sum_{a_c} (a_o - a_c) P(n, a_c; t) (E_n^{-1} - 1) n \\ & + \frac{\mu}{a_o} \sum_n \sum_{a_c} a_c P(n, a_c; t) (E_{a_c}^{-1} - 1) n \\ & + \frac{\lambda}{a_o} \sum_n \sum_{a_c} E_{a_c}^{-1} (a_o - a_c) P(n, a_c; t) E_n^{-1} n \\ & - \frac{\lambda}{a_o} \sum_n \sum_{a_c} (a_o - a_c) P(n, a_c; t) n \end{aligned} \quad (D4)$$

Equation (D3) is derived as follows:

$$(n \in \{0, 1, 2, \dots, N_o\}) \text{ and } (A_c \in \{0, 1, 2, \dots, a_o\})$$

The left-hand side of equation (D3) is

$$\begin{aligned} & \sum_{n=0}^{n_o} \sum_{a_c=0}^{a_o} g(n, a_c) E_n^{-1} E_{a_c}^{-1} f(n, a_c) \\ = & \sum_{n=1}^{n_o+1} \sum_{a_c=1}^{a_o-1} g(n-1, a_c+1) f(n, a_c) \\ = & \sum_{n=1}^{n_o+1} \sum_{a_c=1}^{a_o-1} f(n, a_c) E_n^{-1} E_{a_c}^{-1} g(n, a_c) \end{aligned} \quad (D5)$$

If the two functions  $f(n, a_c)$  and  $g(n, a_c)$  are defined such that the expression being summed up equals zero for

$$n = 0 \text{ and } n_o + 1$$

$$a_c = 0 \text{ and } a_o - 1$$

then equation (D5) is

$$\sum_{n=0}^{n_o} \sum_{a_c=0}^{a_o} g(n, a_c) E_n^{-1} E_{a_c}^{-1} f(n, a_c) = \sum_{n=0}^{n_o} \sum_{a_c=0}^{a_o} f(n, a_c) E_n^{-1} E_{a_c}^{-1} g(n, a_c) \quad (D6)$$

In the case under consideration

$$f(n, a_c) = (a_o - a_c) P(n, a_c; t),$$

and  $g(n, a_c)$  assumes the forms  $n$ ,  $n^2$ ,  $a_c$ ,  $a_c^2$ , or  $na_c$ . In all these instances, the condition stated above holds and hence gives rise to equation (D3).

Equation (D2) is now transformed as

$$\begin{aligned} \frac{d\langle N \rangle}{dt} &= \frac{\kappa}{a_o} \sum_n \sum_{a_c} (a_o - a_c) P(n, a_c; t) (E_n^{-1} - 1)n \\ &+ \frac{\mu}{a_o} \sum_n \sum_{a_c} a_c P(n, a_c; t) (E_{a_c}^{-1} - 1)n \\ &+ \frac{\lambda}{a_o} \sum_n \sum_{a_c} (a_o - a_c) P(n, a_c; t) (E_n^{-1} E_{a_c}^{-1} - 1)n \end{aligned} \quad (D7)$$

which yields the expression for the first moment or expected (mean) value of the random variable  $N$ , denoted by  $\langle N \rangle$ , as

$$\frac{d\langle N \rangle}{dt} = - \frac{(\kappa + \lambda)(a_o - \langle A_c \rangle)}{a_o} \quad (D8)$$

This is equation (34) in the text.

By resorting to similar procedures, the following expressions are obtained for the mean of  $A_c$  and the second moments of  $N$  and  $A_c$  (The second moments of the random variables are denoted by  $\langle N^2 \rangle$  and  $\langle A_c^2 \rangle$  and the crossmoment by  $\langle NA_c \rangle$ ).

$$\frac{d\langle A_c \rangle}{dt} = \frac{\lambda(a_o - \langle A_c \rangle)}{a_o} - \frac{\mu \langle A_c \rangle}{a_o} \quad (D9)$$

$$\frac{d\langle N^2 \rangle}{dt} = - \frac{2(\kappa + \lambda)(a_o \langle N \rangle - \langle NA_c \rangle)}{a_o} + \frac{(\kappa + \lambda)(a_o - \langle A_c \rangle)}{a_o} \quad (D10)$$

$$\begin{aligned} \frac{d\langle A_c^2 \rangle}{dt} = & \frac{\lambda(a_o - \langle A_c \rangle)}{a_o} + \frac{\mu \langle A_c \rangle}{a_o} \\ & + \frac{2\lambda(a_o \langle A_c \rangle - \langle A_c^2 \rangle)}{a_o} - \frac{2\mu \langle A_c^2 \rangle}{a_o} \end{aligned} \quad (D11)$$

$$\begin{aligned} \frac{d\langle NA_c \rangle}{dt} = & - \frac{\kappa(a_o \langle A_c \rangle - \langle A_c^2 \rangle)}{a_o} - \frac{\mu \langle NA_c \rangle}{a_o} \\ & - \frac{\lambda(a_o - \langle A_c \rangle)}{a_o} - \frac{\lambda(a_o \langle A_c \rangle - \langle A_c^2 \rangle)}{a_o} \\ & + \frac{\lambda(a_o \langle N \rangle - \langle NA_c \rangle)}{a_o} \end{aligned} \quad (D12)$$

Equation (D9) is equation (35) in the text.

The variances of  $N$  and  $A_c$  and their covariance are obtained from the above equations based on the following definitions;

$$\langle \langle X \rangle \rangle = \langle X^2 \rangle - \langle X \rangle^2 \quad (D13)$$

$$\langle\langle XY \rangle\rangle = \langle XY \rangle - \langle X \rangle \langle Y \rangle \quad (D14)$$

where  $X$  and  $Y$  are any random variables and  $\langle\langle X \rangle\rangle$  is interpreted as the variance of  $X$ , and  $\langle\langle XY \rangle\rangle$  as the covariance of  $X$  and  $Y$ . The equations for the variances and covariance of  $N$  and  $A_c$  then are:

$$\frac{d\langle\langle N^2 \rangle\rangle}{dt} = 2(\kappa + \lambda) \frac{\langle\langle NA_c \rangle\rangle}{a_o} + (\kappa + \lambda) \left(1 - \frac{\langle A_c \rangle}{a_o}\right) \quad (D15)$$

$$\frac{d\langle\langle A_c^2 \rangle\rangle}{dt} = -2\left(\frac{\lambda + \mu}{a_o}\right) \langle\langle A_c^2 \rangle\rangle + \lambda \left(1 - \frac{\langle A_c \rangle}{a_o}\right) + \mu \frac{\langle A_c \rangle}{a_c} \quad (D16)$$

$$\frac{d\langle\langle NA_c \rangle\rangle}{dt} = (\kappa + \lambda) \frac{\langle\langle A_c^2 \rangle\rangle}{a_o} - \left(\frac{\lambda + \mu}{a_o}\right) \langle\langle NA_c \rangle\rangle - \lambda \left(1 - \frac{\langle A_c \rangle}{a_o}\right) \quad (D17)$$

The initial conditions in this case are expressed as

$$\langle\langle N \rangle\rangle_o = \langle\langle A_c \rangle\rangle_o = \langle\langle NA_c \rangle\rangle_o = 0 \quad (D18)$$

where the subscript  $o$  indicates the reference to time  $t=0$ .

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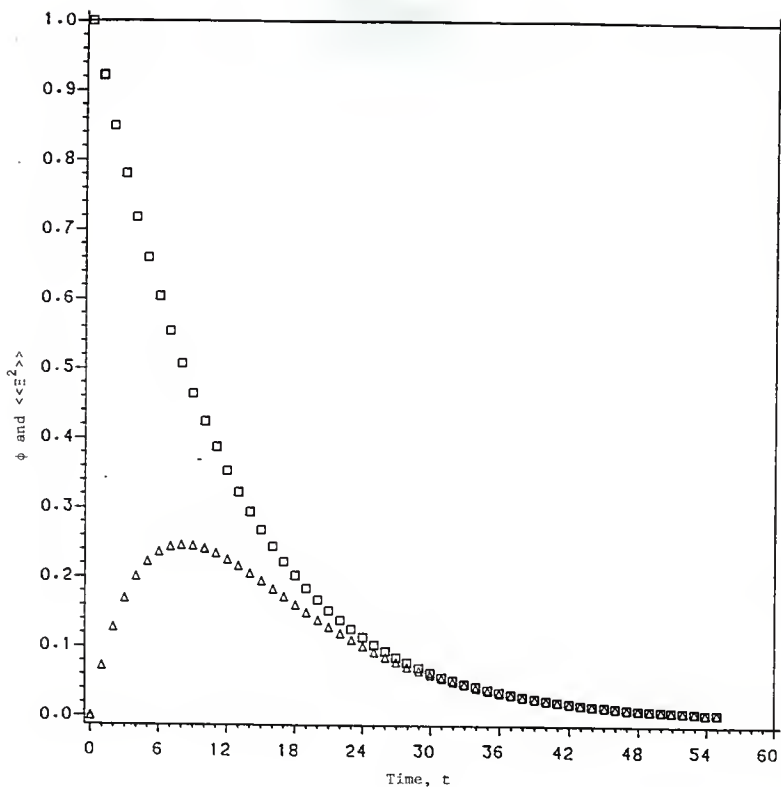


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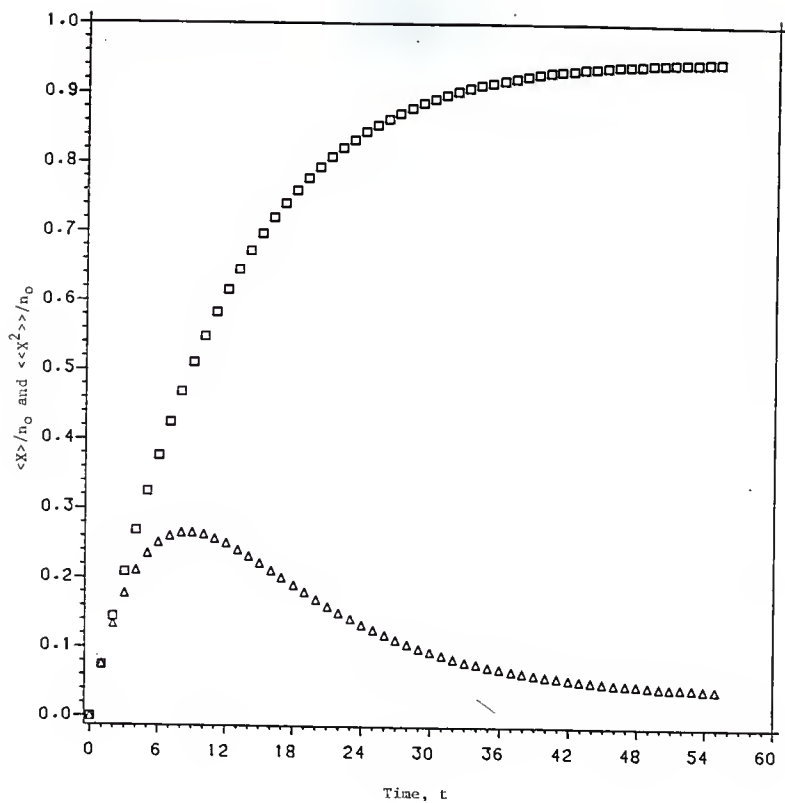


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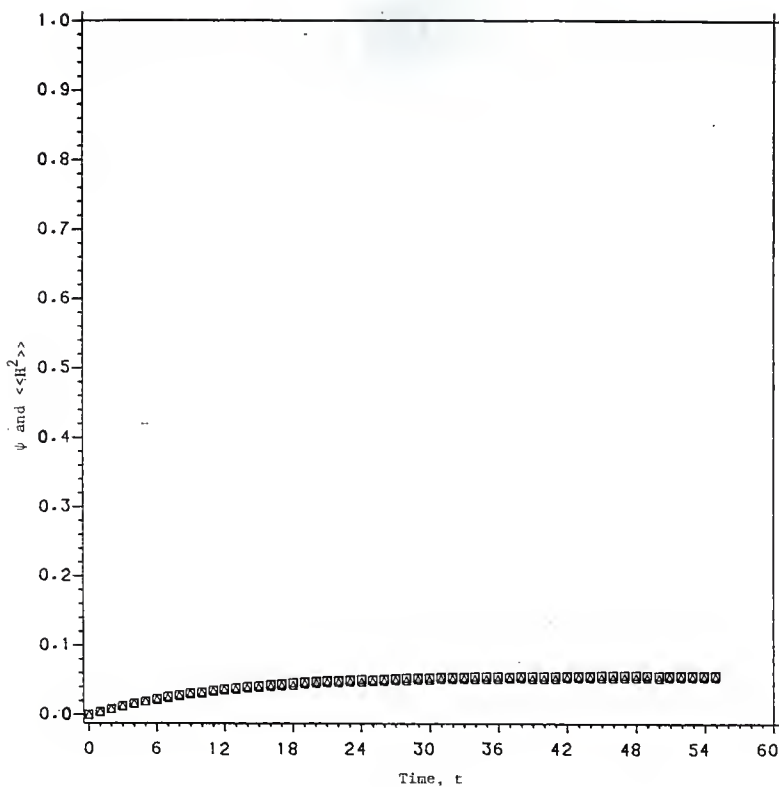


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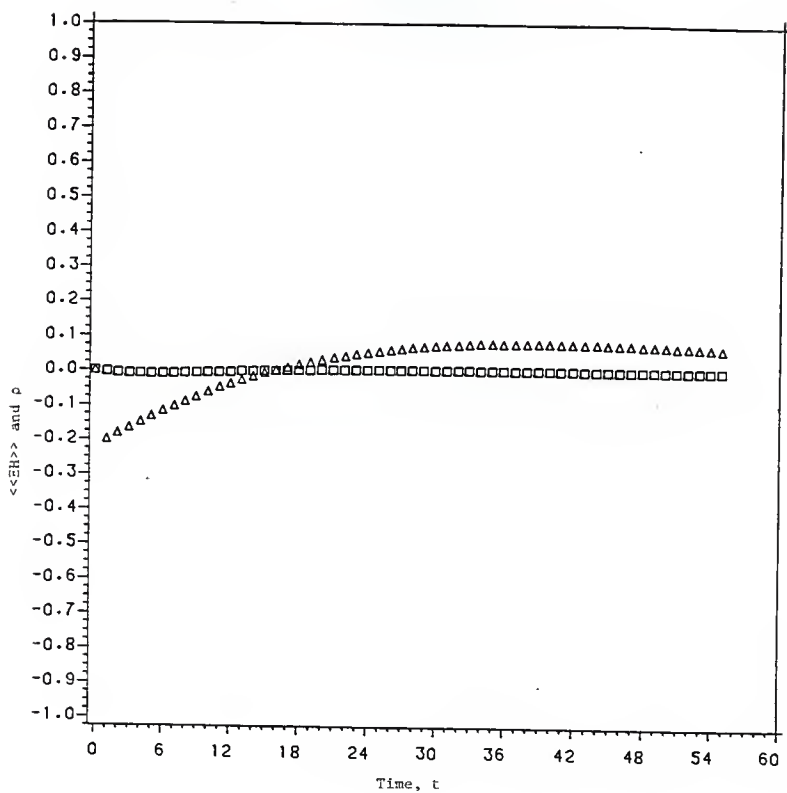


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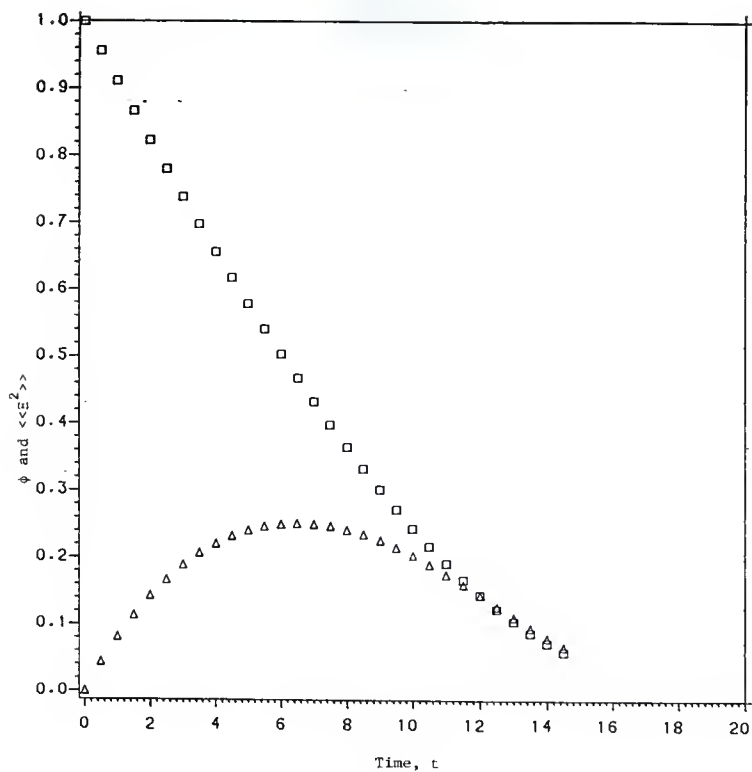


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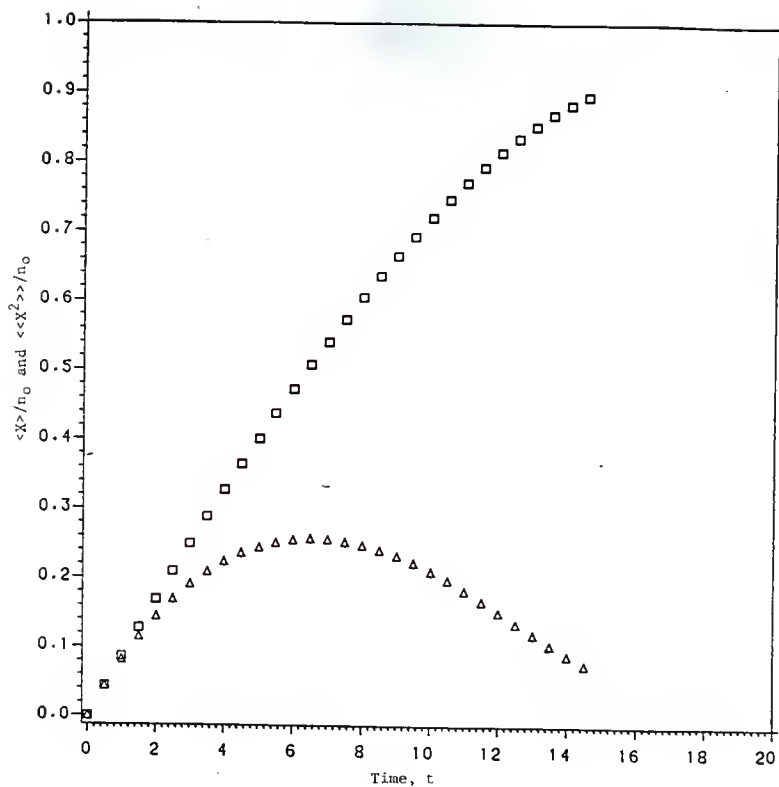


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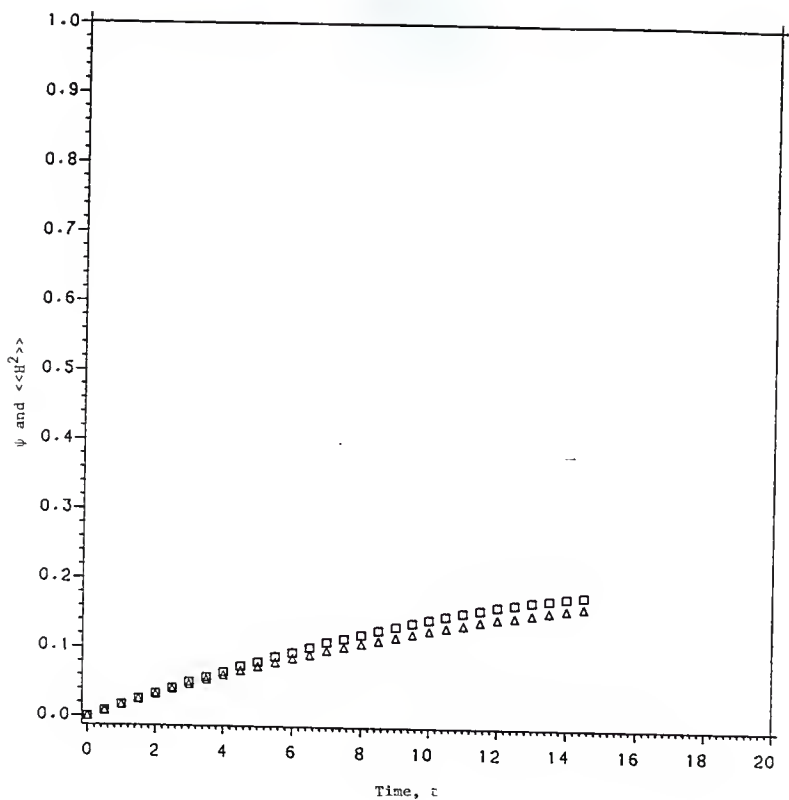


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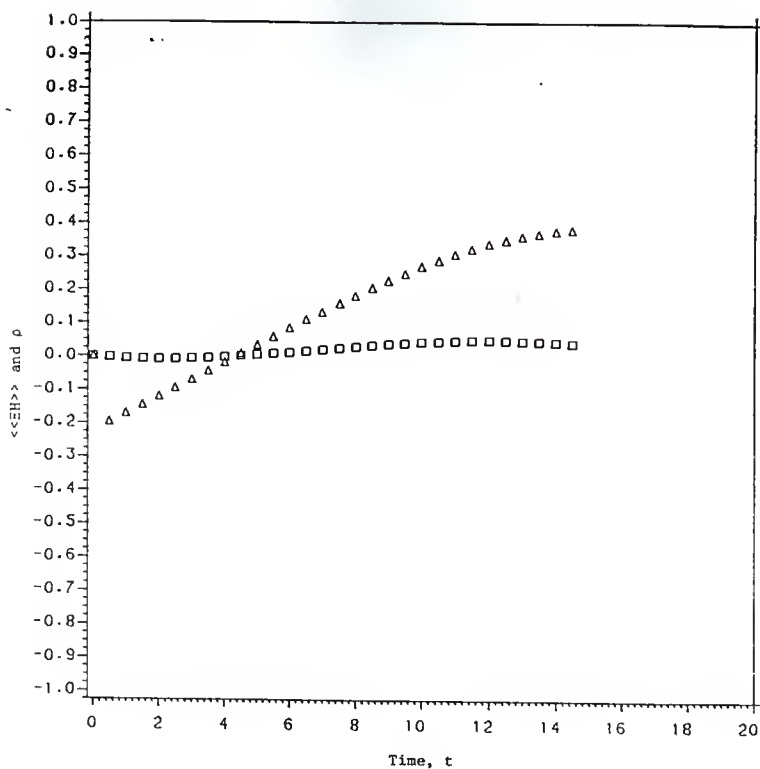


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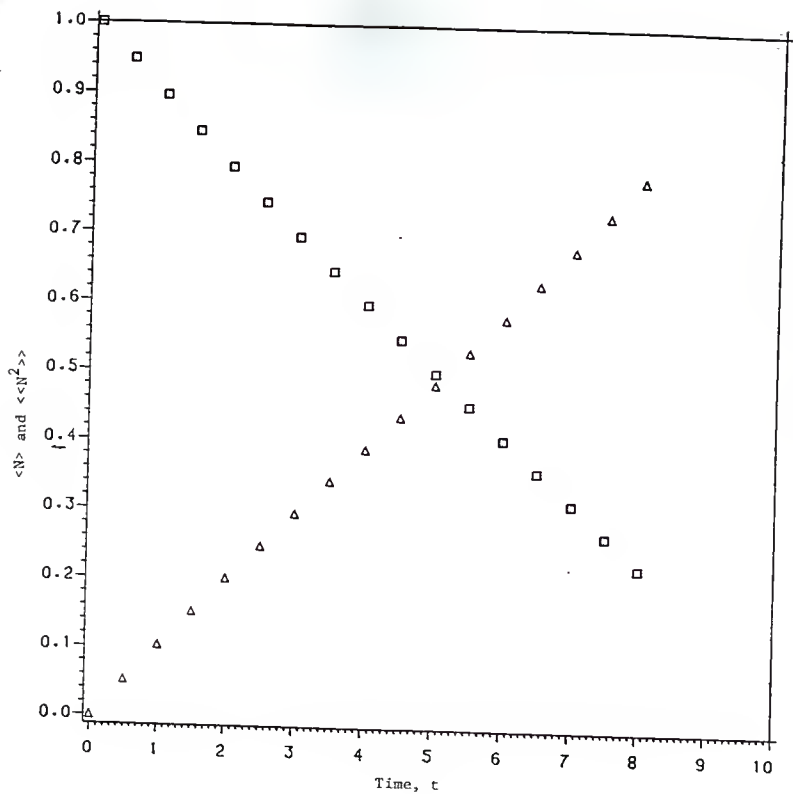


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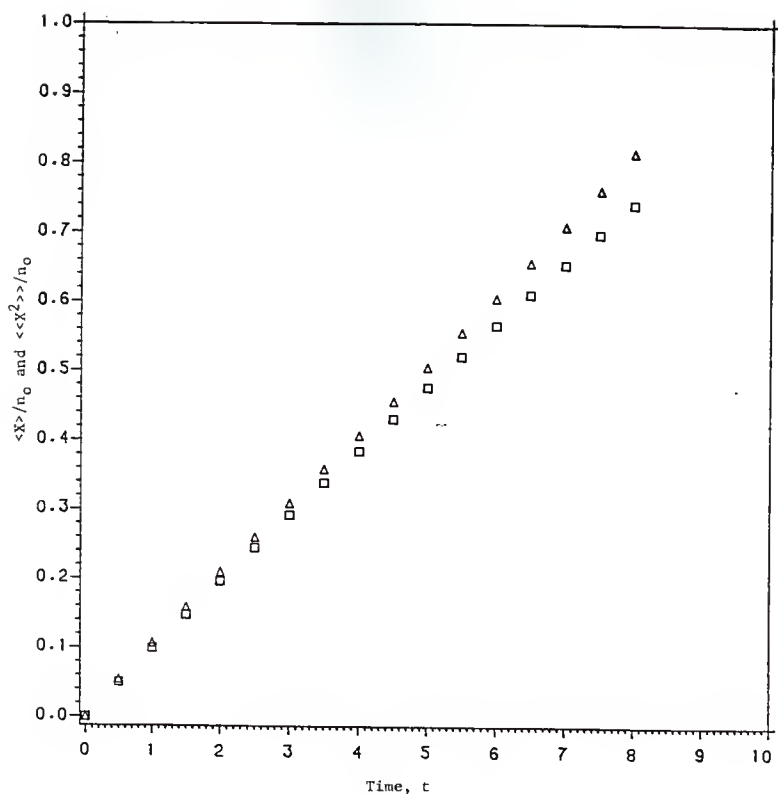


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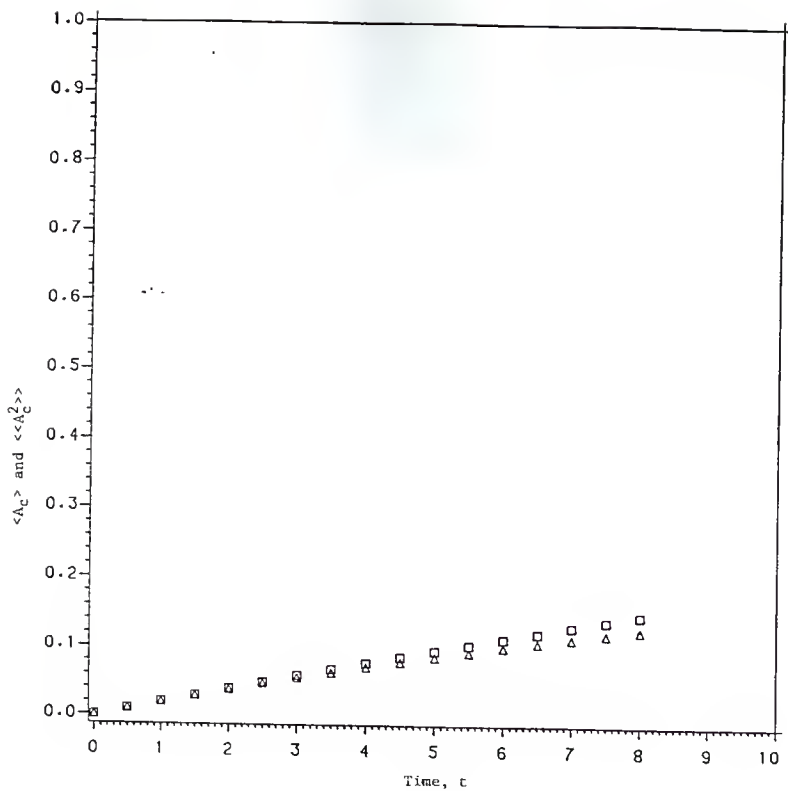


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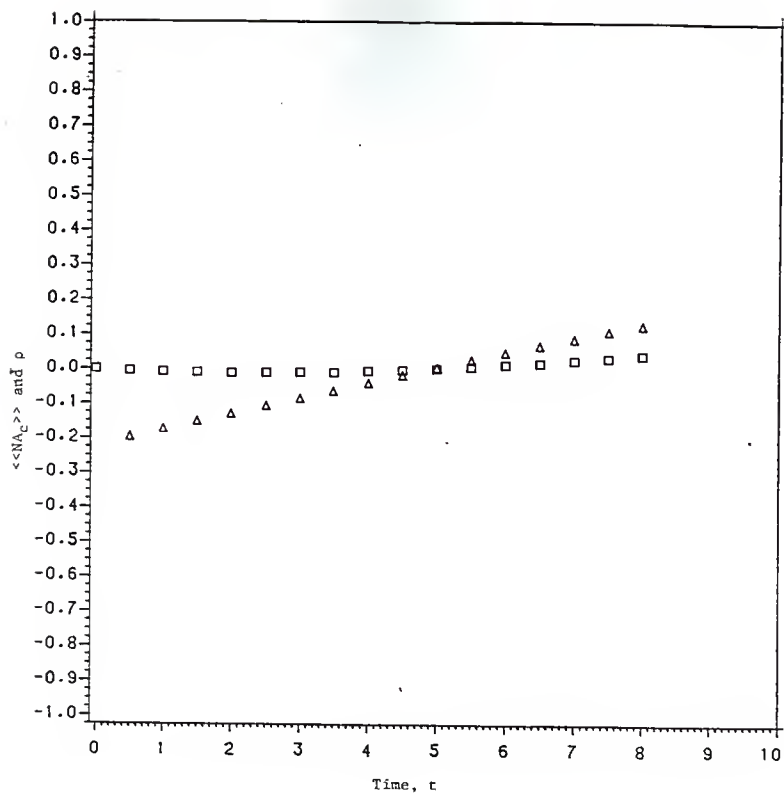


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## CHAPTER 4

### STOCHASTIC ANALYSIS OF COMMINUTION - THE MASTER EQUATION FOR THE GRINDING PROCESS

#### INTROOUCTION

Comminution or size reduction is one of the most commonly used industrial unit operations. The range of applications includes the chemical and mineral processing sectors. Comminution refers simply to the breaking of larger particles into smaller ones. The purpose of comminution may vary depending on the application. For instance, in the mineral processing industry, size reduction is employed to expose the various constituents of rock, thereby facilitating the extraction of valuable minerals through subsequent operations. In chemical processing industry, specifically in solid-liquid extraction or leaching, the particulate material is pulverized to increase the surface area, thereby enhancing the dissolution rate.

Size reduction is commonly achieved by stressing particles to an extent exceeding the tensile strength of the particulate material. The extent and efficiency of the size reduction of solids form the core of industrial mill design. The extent of size reduction depends on the capacity of the mill and has an important bearing on the energy consumption. The relationship between energy input and size reduction can be expressed in a general form as (see, e.g., Weismantel and Sresty, 1985)

$$dE = -C \frac{dx}{x^n} \quad (1)$$

where E is the work done, x is the particle size, and C and n are constants. The familiar Kick's law and Bond's law results from equation (1) for n=1 and n=2 respectively. However, it has been found that, these laws do not yield satisfactory results in practice (see, e.g., Austin and Rogers, 1985).

The extent of size reduction and its efficiency are affected by various factors including the geometry of the mill, the grinding medium, the type of forces acting on the particulate material, and the mechanical properties of the solids. More specifically, the rate of breakage in a size reduction device can be evaluated from the details of (Rogers and Austin, 1985)

1. the stresses on each particle produced by the forces applied during operation,
2. the rate of selection of stressing,
3. the tensile regions in each particle,
4. the fraction of tensile stresses produced that exceed the failure point for the particular flaws in the solids, and
5. crack propagation within a particle.

The complexity involved in describing the particulate system or the size reduction operation on such a microscopic scale demands an alternative means of predicting the results of the operation.

At any instant within the mill, a particle is selected and stressed randomly at various points on its surface and to a degree depending on the magnitude of the forces prevailing in the mill. This may or may not

result in its disintegration. Thus, the cumulative effect of various factors gives rise to a probability that a particle fractures at any given instant.

Attempts have been made in this direction of applying probability theory to explain the internal dynamics of the comminution operation (see, e.g., Hori and Uchida, 1967). Some of these attempts basically recognize the importance of the probabilistic interactions among the particles and between the particles and the grinding media (see, e.g., Rose, 1957). A few probabilistic models have been proposed; which are sketchy and unsatisfactory as they fail to detail the mode of breakage and interactions at the particle level, simultaneously relating to the macroscopic observations. This is important in providing a realistic process description. The macroscopic observations are those that correspond to the results of the well known grinding equation (see, e.g., Rogers and Austin, 1985).

In the present work a description of the grinding process on a mesoscopic level is provided through the formulation of the master equation. Based on a probabilistic consideration of the breakage of an individual particle, this approach gives rise to the macroscopic observations, i.e., the average values of a various particle population sizes, concomitant with certain assumptions. In addition, it yields information on the fluctuations occurring about these mean values. These fluctuations arise from the stochastic nature of the breakage process. The present mesoscopic description of grinding is more fundamental than the conventional grinding equation of the macroscopic type; yet it is not complicated by the details of the microscopic phenomena.

#### DERIVATION OF THE MASTER EQUATION

A particulate process is characterized by the evolution of a population of particles of different characteristics, such as size and shape. The evolution of this population occurs through the events of transformations accomplished by certain external entities, such as the grinding media in ball milling, or interactions between these particles. A stochastic model for the evolution of the particle population can be derived from the concepts of probability theory. The resultant expression for the joint probability of the random variables, representing the distinct groups of particles of the same characteristics, is known as the master equation (see, e.g., van Kampen, 1981 and Gardiner, 1983). An assumption that the events occurring in this population possess the Markov property is essential in formulating the master equation. The Markov property implies that the future events of interactions among particles or their transformations depend solely on the present state of the population and not on its past states.

Let the random variable  $N_i$  represent the number of particles possessing a distinct feature. These groups are distinguished by subscript  $i$ . Let  $s$  represent the total number of such distinct groups existing in the particulate population. The joint probability of the random variables  $\{N\}$  or  $\{N_i; i=1,2,\dots,s\}$  at time  $t$  is denoted as  $P(\{n\},t)$  or simply  $P$ , where  $[\{n\} ; n \in (0,1,2,3,\dots)]$ , i.e., the state space of  $N$  consists of the positive integers. It may be required to approximate  $n$  as a positive real number, i.e.,  $[\{n\} ; n \in (0,+\infty)]$  for the convenience of mathematical manipulation while approximating the



master equation with a Fokker-Planck equation as elaborated later. In such a case, the joint probability  $P$  is the joint probability density function denoted by  $p(\{n\}, t)$ , or simply  $p$ .  $P(\{n\}, t)$  is interpreted as  $P(\{N_1 = n_1, N_2 = n_2, \dots\}, t)$  which is the joint probability that the random variable  $N_1$  has a value  $n_1$ , the random variable  $N_2$  a value of  $n_2$ , and so on, at time  $t$ . The conditional probability,  $P(\{n\}_1, t_1 | \{n\}_0, t_0)$  is the probability that the random variable  $N_1$  has a value of  $n_{11}$ , the random variable  $N_2$  a value of  $n_{21}$ , and so on, at time  $t_1$ , given that the random variable  $N_1$  has a value of  $n_{10}$ , the random variable  $N_2$  has a value of  $n_{20}$ , and so on, at time  $t_0$ .

The quantity  $W_t(\{n\}_0, \{n\}_1)$  is the transition probability per unit time, transition intensity or transition rate of a change in the state of the population from  $\{n\}_0$  to  $\{n\}_1$  in the time interval between  $t$  and  $t+\tau$ . Then  $\tau W_t(\{n\}_0, \{n\}_1)$  is the transition probability from state  $\{n\}_0$  to  $\{n\}_1$  during a small time interval  $\tau$  approaching zero. Also

$$[1 - \tau \sum_{\{n\}_k} W_t(\{n\}_0, \{n\}_k)]$$

is the probability that no transition occurs during the time interval  $\tau$ . Assuming that the various events possess the Markov property,  $P(\{n\}_1, t+\tau)$  is expressed as

$$P(\{n\}_1, t+\tau) = \sum_{\{n\}_k} P(\{n\}_1, t+\tau | \{n\}_k, t) P(\{n\}_k, t) \quad (2)$$

where the subscript  $k$  refers to a particular state of the particle population. The term  $P(\{n\}_1, t + \tau | \{n\}_k, t)$  is expanded in a Taylor series, and taking the limit as  $\tau \rightarrow 0$  yields a generalized form of the master equation (see Fox and Fan, 1985)

$$\frac{dP(\{n\}_1, t)}{dt} = \sum_{\{n\}_k} \{W_t(\{n\}_k, \{n\}_1)P(\{n\}_k, t) - W_t(\{n\}_1, \{n\}_k)P(\{n\}_1, t)\} \quad (3)$$

This expression is a gain-loss equation for the probability of each state  $\{n\}_k$ . The inclusion of the various events and their transition intensities yields the specific master equation for the grinding process.

#### Determination of Transition Intensities for the Grinding Process

Particle disintegration in a mill can occur through three distinct breakage mechanisms (Austin et al., 1986). These are abrasion, breakage, and chipping (the a-b-c of grinding), the first and the last sometimes collectively regarded as attrition. Abrasion is the removal of material from the surface of a particle and can be caused by shear forces acting on the particle. Breakage is the regular fracture of a particle into smaller pieces. It is caused by the specific action of the grinding media on the particulate material, e.g., the crushing of particles by balls in a tumbling mill. Chipping is the conversion of particles which may be highly irregular in shape into rounded particles. This mechanism is usually predominant in the initial stages of the milling operation. The effect of chipping and abrasion is the same in

the sense that both modes of breakage produce a particle of a size close to the original, simultaneously generating fines.

To improve the physical description of the grinding process along with the mathematical procedure, a set of random variables

$$\{N_{i,j}; i \in \{1,2\}, j \in \{1,2,\dots,s\}\}$$

are defined such that  $N_{i,j}$  represents the number of particles of shape  $i$  and of volume  $j\Delta V$  or size  $j$ . Note that these random variables with double subscripts,  $\{N_{i,j}\}$  can be identified with those possessing single subscripts defined earlier,  $\{N_i\}$ , through a transformation from double subscript to single subscript expressed by

$$N_{i+2(j-1)} = N_{i,j} \quad (4)$$

A particle of shape 1 is "stable; it is rounded but can undergo abrasion or breakage. A particle of shape 2 is irregular or in an "excited" state; it can undergo chipping or breakage. Particles from the raw feed or that are freshly broken are considered to be irregular.  $\Delta V$  is the volume of the largest particle classified as a fine particle. The choice of  $\Delta V$  depends on the range of particle sizes being considered. A particle may break into two or more fragments. In this model, however, binary breakage is assumed to predominate. This implies that the probability of more than two particles resulting from a breakage event is low and hence negligible. This is not a serious handicap as it may be possible to include tertiary or higher order breakage by considering an appropriate time interval for the occurrence

of such an event. Multiple breakage can then be represented by a suitable combination of the a-b-c events.

The system considered here is a compartment and the derivation is restricted to a single compartment. The model can be extended to include multi-compartment continuous flow and multiple fragmentation that cannot be represented as mentioned previously.

The a-b-c events are denoted in the following manner:

Abrasion:  $\text{particle}_{1,j} \rightarrow \text{particle}_{1,j-1} + \text{particle}_{1,1}$ ,

$$j > 1$$

Breakage:  $\text{particle}_{1,j} \rightarrow \text{particle}_{2,j-k} + \text{particle}_{2,k}$ ,

$$i = 1, 2; j > 3; 1 < k < (j/2)$$

Chipping:  $\text{particle}_{2,j} \rightarrow \text{particle}_{1,j-1} + \text{particle}_{1,1}$ ,

$$j > 1$$

The subscripts have a similar representation as those of the random variables defined. These events are highly representative of those occurring in a mill and are in accordance with recent investigations (see, e.g., Austin et al., 1986). It is, however, highly improbable that during an abrasion or chipping event, only a single fine is generated. A more realistic description is the generation of fines of a total volume of  $\Delta V$ . Consequently, the random variable  $N_{1,1}$  represents the number of clusters of fines, each cluster having a volume  $\Delta V$ . The assumption that a single stable particle of volume  $\Delta V$  is generated during attrition or chipping, as included in the description of the a-b-c events, does not affect the mean value of fines. However, then, the

variance of  $N_{1,1}$  or the covariances involving  $N_{1,1}$  do not truly characterize the number of fines. Therefore, letting  $N_{1,1}$  represent the number of clusters of fines is appropriate when considering that the fines do not undergo any of the a-b-c events, and assuming that each clusters of volume  $\Delta V$  behaves as a single entity.

Letting  $\alpha_j$  represent the specific rate of abrasion of size  $j$  particles into size  $j-1$  particles, the transition rate of abrasion can be written as

$$W_t(\{n_{1,j}, n_{1,j-1}, n_{1,1}\}, \{n_{1,j-1}, n_{1,j-1}+1, n_{1,1}+1\}) \\ = \alpha_j n_{1,j}, \quad j = 2, 3, \dots, s \quad (5)$$

The specific rate of breakage depends on the size being broken as well as the sizes of the daughter fragments. This is represented by the specific rate function for breakage  $\beta_{i,j}(k)$ . Consequently the transition intensity of breakage is expressed as

$$W_t(\{n_{1,j}, n_{2,j-k}, n_{2,k}\}, \{n_{1,j-1}, n_{2,j-k}+1, n_{2,k}+1\}) \\ = \beta_{1,j}(k) n_{1,j}, \quad i = 1, 2; \quad j = 4, 5, \dots, s, \quad j \neq 2k; \quad 1 < k < (\frac{j}{2}) \quad (6) \\ W_t(\{n_{1,j}, n_{2,k}\}, \{n_{1,j-1}, n_{2,k}+2\}) \\ = \beta_{i,j}(k) n_{1,j}, \quad i = 1, 2; \quad j = 4, 5, \dots, s; \quad j = 2k \quad (7)$$

Similarly, letting  $\nu_j$  represent the specific rate of chipping, the transition rate of chipping is written as

$$W_t(\{n_{2,j}, n_{1,j-1}, n_{1,1}\}, \{n_{2,j-1}, n_{1,j-1}+1, n_{1,1}+1\})$$

$$= v_j n_{2,j}, \quad j = 2, 3, \dots, s \quad (8)$$

For simplicity, all random variables remaining constant during a transition have not been included in the notation for  $\{n\}$  in equations (5) through (8). It can be seen from these equations that the transition intensities of the a-b-c events are proportional to the number of particles. In general the intensities of abrasion and chipping given by equations (5) and (8), respectively are functions of initial sizes. The intensity of breakage given by equations (6) and (7) is a function of the daughter particle size in addition to the initial size. Note that the linear dependence on the number may not be valid for all modes of grinding. For instance, interparticle collisions largely effect size reduction through breakage, in autogeneous grinding. In this case, a nonlinear dependence of the transition intensity of breakage may be encountered. Furthermore, the presence of fines may reduce the rates of abrasion, breakage and chipping. This would necessitate the inclusion of the number of fines in the expressions for the transition intensities. This difficulty can be circumvented by rendering the parameters  $\alpha_j$ ,  $\beta_{i,j}$ , and  $v_j$  in equations (5) through (8) as some functions of time.

Letting  $\lambda_{i,j}$  represent the rate of inflow of particles of shape  $i$  and size  $j$  to the compartment. Then, it is reasonable to assume that the transition intensity for the entrance of particles is

$$W_t(\{n_{i,j}\}, \{n_{i,j}+1\}) = \lambda_{i,j}, \quad i = 1, 2; \quad j = 1, 2, \dots, s \quad (9)$$

The rate at which particles exit is represented as  $\mu_{i,j}$ . The corresponding exit transition intensity can be assumed to have the form

$$w_c(\{n_{i,j}\}, \{n_{i,j}-1\}) = \mu_{i,j} n_{i,j}, \quad i = 1, 2; \quad j = 1, 2, \dots, s \quad (10)$$

The transition intensities can be written given by eqs. (5) through (10) in terms of the values assumed by the random variables with single subscript i.e.,  $\{N_i\}$ . Their subsequent inclusion in equation (3) yields the master equation for the grinding process.

It is often difficult to write the master equation explicitly by specifying the events and their transition intensities. Besides it is extremely difficult, if not impossible to solve exactly for the joint probability distribution. This holds for the moments of the distribution except for a few simple instances, albeit to a much lesser degree. In fact it is often more useful to know the moments of the probability distribution rather than the distribution itself. Methods are available to solve the master equation depending on its complexity. When the transition intensities are simple linear functions of the random variables and the number of variables small, the probability generating functions technique can be employed to arrive at a set of differential equations for the moments and in some cases, a form of the probability distribution itself (see, e.g., Oppenheim et al., 1977). A direct solution can also be obtained by using the definition of the expected value of a function of a random variable. If  $X$  is a random variable and  $g(X)$  a function of the random variable, the expected value of  $g(X)$  denoted by  $E[g(X)]$ , is defined as

$$E[g(X)] = \sum_j g(x_j) f_X(x_j) \quad (11)$$

where  $X$  is discrete with possible values  $x_1, x_2, \dots, x_j, \dots$ . Statistical quantities such as the means and variances can be derived by a suitable selection of  $g(X)$ . If the number of variables are large and the transition intensities, non-linear functions of the random variables, then a rational approximation procedure such as the system size expansion is required to solve the master equation (see, e.g., van Kampen, 1981, and Fox and Fan, 1985).

#### APPROXIMATION OF THE MASTER EQUATION

In the present work the generalized master equation, equation (3) is approximated by a Fokker-Planck equation. In doing so, it is assumed that the transition intensities are smooth functions of the random variables and that the probability distribution  $P(\{n\}, t)$  can be represented by a smooth function  $p(\{n\}, t)$ , the probability density function. Also, the individual step changes in the random variables are assumed to be small in comparison with the magnitude of the random variables themselves. Hence, such an approximation is reasonable. In fact this procedure yields exact macroscopic equations and equations for the second moments in such cases involving simple linear functions for the transition intensities. However, the corresponding equations for the higher moments of the probability distribution may not be correctly reproduced by the Fokker-Planck equation (see, e.g., van Kampen, 1981). This is of minor importance as these moments are usually physically insignificant.



The transition intensities to be included in the master equation, equation (3) are now rewritten in terms of the reference vector,  $\{n\}_k$ , and the magnitude of the change in the random vector,  $\{n\}_1 - \{n\}_k$ . This facilitates the expansion of the master equation to form the Fokker-Planck equation. Letting  $\{\xi\}_k = \{n\}_1 - \{n\}_k$ , the transition intensities are expressed as

$$W_t(\{n\}_k; \{n\}_1) = W_t(\{n\}_1 - \{\xi\}_k; \{\xi\}_k) \quad (12)$$

where  $\{\xi\}_k$  is the vector whose components are  $\xi_{ik}$ , i.e.,

$$\{\xi\}_k = \{\xi_{1k}, \xi_{2k}, \dots, \xi_{ik}, \dots\}$$

The subscript  $k$  indicates the state of the population. Consequently the master equation assumes the form

$$\begin{aligned} \frac{dP(\{n\}_1, t)}{dt} = \sum_{\{\xi\}_k} [W_t(\{n\}_1 - \{\xi\}_k; \{\xi\}_k) P(\{n\}_1 - \{\xi\}_k, t) \\ - W_t(\{n\}_1; \{\xi\}_k) P(\{n\}_1, t)] \end{aligned} \quad (13)$$

The transition intensities for the grinding process are written in terms of  $\{\xi\}$  as

$$\begin{aligned} W_t(\{n_{1,j}, n_{1,j+1}, n_{1,1}\}, \{-1, +1, +1\}) = \alpha_j n_{i,j}, \\ j = 2, 3, \dots, s \end{aligned} \quad (14)$$

$$\begin{aligned} W_t(\{n_{i,j}, n_{2,j-k}, n_{2,k}\}, \{-1, +1, +1\}) = \beta_{i,j}(k) n_{i,j}, \\ i = 1, 2; j = 4, 5, \dots, s; j \neq 2k \end{aligned} \quad (15)$$

$$\begin{aligned} W_t(\{n_{i,j}, n_{2,k}\}, \{-1, +2\}) = \beta_{i,j}(k) n_{i,j}, \\ i = 1, 2; j = 2k \end{aligned} \quad (16)$$

$$W_t(\{n_{2,j}, n_{1,j-1}, n_{1,1}\}, \{-1, +1, +1\}) = \nu_j n_{2,j},$$

$$j = 2, 3, \dots, s \quad (17)$$

$$W_t(\{n_{i,j}\}, \{+1\}) = \lambda_{i,j},$$

$$i = 1, 2; j = 1, 2, \dots, s \quad (18)$$

$$W_t(\{n_{i,j}\}, \{-1\}) = \mu_{i,j} n_{i,j},$$

$$i = 1, 2; j = 1, 2, \dots, s \quad (19)$$

The Taylor expansion of the first term on the right hand side of equation (13) around  $\{n\}_1$  yields (see, e.g., Fox and Fan, 1985)

$$\begin{aligned} & \frac{\partial p(\{n\}_1, t)}{\partial t} \\ &= \sum_{\{\xi\}_k} [W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t) - W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t)] \\ & \quad - \sum_{\{\xi\}_k} \sum_i \xi_{ik} \frac{\partial}{\partial n_{i1}} [W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t)] \\ & \quad + \frac{1}{2} \sum_{\{\xi\}_k} \sum_i \sum_j \xi_{ik} \xi_{jk} \frac{\partial^2}{\partial n_{i1} \partial n_{j1}} [W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t)] + o(\xi^3) \\ &= - \sum_i \frac{\partial}{\partial n_{i1}} \left[ \sum_{\{\xi\}_k} \xi_{ik} W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t) \right] \\ & \quad + \frac{1}{2} \sum_i \sum_j \frac{\partial^2}{\partial n_{i1} \partial n_{j1}} \left[ \sum_{\{\xi\}_k} \xi_{ik} \xi_{jk} W_t(\{n\}_1; \{\xi\}_k) p(\{n\}_1, t) \right] + o(\xi^3) \quad (20) \end{aligned}$$

where  $\xi_{ik}$  and  $\xi_{jk}$  are the components of  $\{\xi\}_k$ , i.e., the changes in the magnitudes of the variables  $N_i$  and  $N_j$  corresponding to state  $k$  of the particle population, i.e.,  $\{n\}_k$ . Equation (20) is equally valid for any other state as it is for  $\{n\}_1$ . Hence, it is written in a general form as

$$\begin{aligned} \frac{\partial p(\{n\}, t)}{\partial t} = & - \sum_k \sum_i \frac{\partial \{A_{i,k}(\{n\}, t) p(\{n\}, t)\}}{\partial n_i} \\ & + \frac{1}{2} \sum_j \sum_i \frac{\partial^2 \{B_{i,j}(\{n\}, t) p(\{n\}, t)\}}{\partial n_i \partial n_j} \end{aligned} \quad (21)$$

This is a multivariate Fokker-Planck equation with coefficients  $A_{i,k}(\{n\}, t)$  and  $B_{i,j}(\{n\}, t)$  defined as

$$\begin{aligned} A_{i,k}(\{n\}, t) &= (n_{ik} - n_i) W_t(\{n\}, \{n\}_k) \\ &= \xi_{ik} W_t(\{n\}, \{\xi\}_k) \end{aligned} \quad (22)$$

$$\begin{aligned} B_{i,j}(\{n\}, t) &= \sum_k (n_{ik} - n_i)(n_{jk} - n_j) W_t(\{n\}, \{n\}_k) \\ &= \sum_k \xi_{ik} \xi_{jk} W_t(\{n\}; \xi_{ik} \xi_{jk}) \end{aligned} \quad (23)$$

The moments of  $p(\{n\}, t)$  can be obtained through familiar methods (see, e.g., van Kampen, 1981). Letting  $\langle * \rangle$  denote the first moment of random variable  $*$  and  $\langle *+ \rangle$  the cross moment of random variables  $*$  and  $+$ , the expressions for the moments are written as

$$\frac{d\langle N_i \rangle}{dt} = \sum_k \langle A_{i,k}(\{n\}, t) \rangle \quad (24)$$

$$\frac{d\langle N_i N_j \rangle}{dt} = \sum_k \{ \langle A_{i,k} N_j \rangle + \langle A_{j,k} N_i \rangle + \langle B_{i,j}(\{n\}, t) \rangle \} \quad (25)$$

The master equation for the grinding process, given in terms of  $\{N_{i,j}\}$  is solved by this procedure of approximation with the Fokker Planck equation, except that the variables  $\{N_i\}$  are now identified with  $\{N_{i,j}\}$  through the subscript transformation indicated by equation (4).

# NUMERICAL EXAMPLE

Consider a compartment representing a section of a ball mill. The particles in the compartment are assumed to be classified into four size ranges denoted 1 through 4 in ascending order of size with the feed consisting of irregular particles of size 4. Initially a certain number of irregular particles of size 4 exist in the compartment. Corresponding to this description, eight random variables  $N_{2,4}, N_{1,4}, N_{2,3}, N_{1,3}, N_{2,2}, N_{1,2}, N_{2,1}$  and  $N_{1,1}$  represent the numbers of particles of the different shapes and sizes present in the mill compartment.

It can be seen that no contributions to or from sizes  $N_{2,4}, N_{2,3}$  and  $N_{2,1}$  result from the occurrence of the a-b-c events. The unification of the double subscript is then performed simply as

$$N_{2,4} = N_1; N_{1,3} = N_2; N_{2,2} = N_3; N_{1,2} = N_4; N_{1,1} = N_5$$

ignoring  $N_{1,4}, N_{2,3}$  and  $N_{2,1}$ . In doing so the scheme provided in earlier sections is disregarded.

The events and their transition intensities are described in Table 1. These are in accordance with the grinding mechanism detailed earlier. The calculation of the coefficients of the Fokker-Planck equation is illustrated (APPENDIX A). They are presented in terms of the transition intensities in Table 2. The following are the first moments or the macroscopic equations in terms of  $\{N_{i,j}\}$ :

$$\frac{d\langle N_{2,4} \rangle}{dt} = \lambda_{2,4} - (\beta_{2,4}(2) + \nu_4, \mu_{2,4}) \langle N_{2,4} \rangle \quad (26)$$

$$\frac{d\langle N_{1,3} \rangle}{dt} = v_4 \langle N_{2,4} \rangle - (\alpha_3 + \mu_{1,3}) \langle N_{1,3} \rangle \quad (27)$$

$$\frac{d\langle N_{2,2} \rangle}{dt} = 2\beta_{2,4}(2) \langle N_{2,4} \rangle - (v_2 + \mu_{2,2}) \langle N_{2,2} \rangle \quad (28)$$

$$\frac{d\langle N_{1,2} \rangle}{dt} = \alpha_3 \langle N_{1,3} \rangle - (\alpha_2 + \mu_{1,2}) \langle N_{1,2} \rangle \quad (29)$$

$$\begin{aligned} \frac{d\langle N_{1,1} \rangle}{dt} = & v_4 \langle N_{2,4} \rangle + \alpha_3 \langle N_{1,3} \rangle + 2v_2 \langle N_{2,2} \rangle \\ & + 2\alpha_2 \langle N_{1,2} \rangle - \mu_{1,1} \langle N_{1,1} \rangle \end{aligned} \quad (30)$$

The initial conditions at  $t = 0$  are

$$\langle N_{2,4} \rangle = n_0; \quad \langle N_{1,3} \rangle = \langle N_{2,2} \rangle = \langle N_{1,2} \rangle = \langle N_{1,1} \rangle = 0$$

Instead of arriving at the equations for the cross moments, the covariances can be directly calculated using the definition of the covariance of two random variables X and Y given as

$$\langle \langle XY \rangle \rangle = \langle XY \rangle - \langle X \rangle \langle Y \rangle$$

This gives rise to the following equations result for the covariances:

$$\begin{aligned} \frac{d\langle \langle N_{2,4}^2 \rangle \rangle}{dt} = & -2(\beta_{2,4}(2) + v_4 + \mu_{2,4}) \langle \langle N_{2,4}^2 \rangle \rangle \\ & + \lambda_{2,4} + (\beta_{2,4}(2) + v_4 + \mu_{2,4}) \langle N_{2,4} \rangle \end{aligned} \quad (31)$$

$$\begin{aligned} \frac{d\langle \langle N_{2,4} N_{1,3} \rangle \rangle}{dt} = & -(\beta_{2,4}(2) + v_4 + \mu_{2,4}) \langle \langle N_{2,4} N_{1,3} \rangle \rangle \\ & + v_4 \langle \langle N_{2,4}^2 \rangle \rangle - (\alpha_3 + \mu_{1,3}) \langle \langle N_{2,4} N_{1,3} \rangle \rangle \\ & - v_4 \langle N_{2,4} \rangle \end{aligned} \quad (32)$$

$$\frac{d\langle \langle N_{2,4} N_{2,2} \rangle \rangle}{dt} = -(\beta_{2,4}(2) + v_4 + \mu_{2,4}) \langle \langle N_{2,4} N_{2,2} \rangle \rangle$$

$$\begin{aligned}
& + 2\beta_{2,4}(2) \langle\langle N_{2,4}^2 \rangle\rangle - (u_2 + \mu_{2,2}) \langle\langle N_{2,4} N_{2,2} \rangle\rangle \\
& - 2\beta_{2,4}(2) \langle N_{2,4} \rangle
\end{aligned} \tag{33}$$

$$\begin{aligned}
\frac{d\langle\langle N_{2,4} N_{1,2} \rangle\rangle}{dt} &= -(\beta_{2,4}(2) + u_4 + \mu_{2,4}) \langle\langle N_{2,4} N_{1,2} \rangle\rangle \\
&+ \alpha_3 \langle\langle N_{2,4} N_{1,3} \rangle\rangle - (\alpha_2 + \mu_{1,2}) \langle\langle N_{2,4} N_{1,2} \rangle\rangle
\end{aligned} \tag{34}$$

$$\begin{aligned}
\frac{d\langle\langle N_{2,4} N_{1,1} \rangle\rangle}{dt} &= -(\beta_{2,4}(2) + u_4 + \mu_{2,4}) \langle\langle N_{2,4} N_{1,1} \rangle\rangle \\
&+ u_4 \langle\langle N_{2,4}^2 \rangle\rangle + \alpha_3 \langle\langle N_{2,4} N_{1,3} \rangle\rangle \\
&+ 2u_2 \langle\langle N_{2,4} N_{2,2} \rangle\rangle + 2\alpha_2 \langle\langle N_{2,4} N_{1,2} \rangle\rangle \\
&- \mu_{1,1} \langle\langle N_{2,4} N_{1,1} \rangle\rangle - u_4 \langle N_{2,4} \rangle
\end{aligned} \tag{35}$$

$$\begin{aligned}
\frac{d\langle\langle N_{1,3}^2 \rangle\rangle}{dt} &= 2u_4 \langle\langle N_{2,4} N_{1,3} \rangle\rangle - 2(\alpha_3 + \mu_{1,3}) \langle\langle N_{1,3}^2 \rangle\rangle \\
&+ (\alpha_3 + \mu_{1,3}) \langle N_{1,3} \rangle + u_4 \langle N_{2,4} \rangle
\end{aligned} \tag{36}$$

$$\begin{aligned}
\frac{d\langle\langle N_{1,3} N_{2,2} \rangle\rangle}{dt} &= u_4 \langle\langle N_{2,4} N_{2,2} \rangle\rangle + 2\beta_{2,4}(2) \langle\langle N_{2,4} N_{1,3} \rangle\rangle \\
&- (\alpha_3 + \mu_{1,3}) \langle\langle N_{1,3} N_{2,2} \rangle\rangle \\
&- (u_2 + \mu_{2,2}) \langle\langle N_{1,3} N_{2,2} \rangle\rangle
\end{aligned} \tag{37}$$

$$\begin{aligned}
\frac{d\langle\langle N_{1,3} N_{1,2} \rangle\rangle}{dt} &= u_4 \langle\langle N_{2,4} N_{1,2} \rangle\rangle - (\alpha_3 + \mu_{1,3}) \langle\langle N_{1,3} N_{1,2} \rangle\rangle \\
&+ \alpha_3 \langle\langle N_{1,3}^2 \rangle\rangle - (\alpha_2 + \mu_{1,2}) \langle\langle N_{1,3} N_{1,2} \rangle\rangle \\
&- \alpha_3 \langle N_{1,3} \rangle
\end{aligned} \tag{38}$$

$$\frac{d\langle\langle N_{1,3} N_{1,1} \rangle\rangle}{dt} = u_4 \langle\langle N_{2,4} N_{1,1} \rangle\rangle + u_4 \langle\langle N_{2,4} N_{1,3} \rangle\rangle$$

$$\begin{aligned}
& - (\alpha_3 + \mu_{1,3}) \langle N_{1,3} N_{1,1} \rangle + \alpha_3 \langle N_{1,3}^2 \rangle \\
& + 2v_2 \langle N_{1,3} N_{2,2} \rangle + 2\alpha_2 \langle N_{1,3} N_{1,2} \rangle \\
& - \mu_{1,1} \langle N_{1,3} N_{1,1} \rangle \\
& - \alpha_3 \langle N_{1,3} \rangle + v_4 \langle N_{2,4} \rangle
\end{aligned} \tag{39}$$

$$\begin{aligned}
\frac{d \langle N_{2,2}^2 \rangle}{dt} &= 4\beta_{2,4} \langle N_{2,4} N_{2,2} \rangle - 2(v_2 + \mu_{2,2}) \langle N_{2,2}^2 \rangle \\
&+ 4\beta_{2,4} \langle N_{2,4} \rangle + (v_2 + \mu_{2,2}) \langle N_{2,2} \rangle
\end{aligned} \tag{40}$$

$$\begin{aligned}
\frac{d \langle N_{2,2} N_{1,2} \rangle}{dt} &= 2\beta_{2,4} \langle N_{2,4} N_{1,2} \rangle - (v_2 + \mu_{2,2}) \langle N_{2,2} N_{1,2} \rangle \\
&+ \alpha_3 \langle N_{1,3} N_{2,2} \rangle - (\alpha_2 + \mu_{1,2}) \langle N_{2,2} N_{1,2} \rangle
\end{aligned} \tag{41}$$

$$\begin{aligned}
\frac{d \langle N_{2,2} N_{1,1} \rangle}{dt} &= 2\beta_{2,4} \langle N_{2,4} N_{1,1} \rangle - (v_2 + \mu_{2,2}) \langle N_{2,2} N_{1,1} \rangle \\
&+ v_4 \langle N_{2,4} N_{2,2} \rangle + \alpha_3 \langle N_{1,3} N_{2,2} \rangle \\
&+ 2v_2 \langle N_{2,2}^2 \rangle + 2\alpha_2 \langle N_{2,2} N_{1,2} \rangle \\
&- \mu_{1,1} \langle N_{2,2} N_{1,1} \rangle - 2v_2 \langle N_{2,2} \rangle
\end{aligned} \tag{42}$$

$$\begin{aligned}
\frac{d \langle N_{1,2}^2 \rangle}{dt} &= 2\alpha_3 \langle N_{1,3} N_{1,2} \rangle - 2(\alpha_2 + \mu_{1,2}) \langle N_{1,2}^2 \rangle \\
&+ \alpha_3 \langle N_{1,3} \rangle + \alpha_2 \langle N_{1,2} \rangle + \mu_{1,2} \langle N_{1,2} \rangle
\end{aligned} \tag{43}$$

$$\begin{aligned}
\frac{d \langle N_{1,2} N_{1,1} \rangle}{dt} &= \alpha_3 \langle N_{1,3} N_{1,1} \rangle - (\alpha_2 + \mu_{1,2}) \langle N_{1,2} N_{1,1} \rangle \\
&+ v_4 \langle N_{2,4} N_{1,2} \rangle + \alpha_3 \langle N_{1,3} N_{1,2} \rangle \\
&+ 2v_2 \langle N_{2,2} N_{1,2} \rangle + 2\alpha_2 \langle N_{1,2}^2 \rangle \\
&- \mu_{1,1} \langle N_{1,2} N_{1,1} \rangle + \alpha_3 \langle N_{1,3} \rangle
\end{aligned}$$

$$-2\alpha_2 \langle N_{1,2} \rangle \quad (44)$$

$$\begin{aligned} \frac{d\langle N_{1,1}^2 \rangle}{dt} = & 2u_4 \langle N_{2,4} N_{1,1} \rangle + 2\alpha_3 \langle N_{1,3} N_{1,1} \rangle \\ & + 4u_2 \langle N_{2,2} N_{1,1} \rangle + 4\alpha_2 \langle N_{1,2} N_{1,1} \rangle \\ & - 2\mu_{1,1} \langle N_{1,1}^2 \rangle + \alpha_3 \langle N_{1,3} \rangle \\ & - 4\alpha_2 \langle N_{1,2} \rangle + u_4 \langle N_{2,4} \rangle \\ & + 4u_2 \langle N_{2,2} \rangle + \mu_{1,1} \langle N_{1,1} \rangle \end{aligned} \quad (45)$$

Table 3 lists the parameter values used in the example calculation. This system of coupled linear ordinary differential equations is solved by resorting to a fourth order Runge-Kutta algorithm.

#### DISCUSSION

The mean values and the variances of the numbers of particles of various sizes and shapes are presented in Figures 1 through 5. The variance of  $N_{1,1}$  is high as expected due to the cumulative effect of the generation of fines from the different sources. The covariances of  $N_{2,4}$  and the other variables are presented as correlation coefficients in Figure 6. The correlation coefficient of two random variables X and Y, denoted as  $\rho(x,y)$  is the ratio of their covariance to the product of their standard deviations. The standard deviation is defined as the square root of the variance. Thus,

$$\rho(X,Y) = \frac{\langle XY \rangle}{\sqrt{\langle X^2 \rangle \langle Y^2 \rangle}} \quad (46)$$



Both the covariance and correlation coefficient of two random variable X and Y are measures of a linear relationship in the sense that the covariance,  $\langle XY \rangle$  and the correlation coefficient (X,Y) will be positive when  $X - \langle X \rangle$  and  $Y - \langle Y \rangle$  tend to have the same sign with high probability. The covariance is negative when  $X - \langle X \rangle$  and  $Y - \langle Y \rangle$  tend to have opposite signs with high probability. The actual value of the covariance is not physically significant as it depends on the individual variability of X and Y. The correlation coefficient removes this individual variability and is hence a better measure of the linear relationship of X and Y. The value of the correlation coefficient must lie between -1 and +1 (Mood et al., 1974).

The correlation coefficients presented in Figure 6 all assume values equal to or below zero. This is because an increased value of  $N_{2,4}$  over its mean indicates that less number of size 4, irregular particles have undergone size reduction. Consequently the values of the other variables will fall below their mean. This holds also for variables indirectly related such as  $N_{2,4}$  and  $N_{1,2}$ , related through  $N_{1,3}$  and  $N_{2,2}$ . The correlation coefficients of other pairs of random variables are presented in Figures 7 through 9. Note that  $\rho(N_{1,3}, N_{1,1})$  in Figure 8 is positive. This is because an increased value of  $N_{1,3}$  above its mean would mean less number of fines clusters generated but it also implies that more size 4, regular particles undergo chipping. The chipping rate of size 4 particles is higher than the abrasion rate of size 3 particles as listed in Table 3, implying a net increase in the

value of  $N_{1,1}$  above its mean. In general, the covariance and the correlation coefficient depend on the transition intensities. Thus, they reflect the different size reduction mechanisms and their rates and are therefore useful in obtaining a qualitative picture of the evolution of the particle population.

## CONCLUDING REMARKS

The analysis of the grinding process on a mesoscopic level, based on probabilistic considerations and the a-b-c grinding mechanisms, definitely provides a more detailed picture than conventional approaches. Efforts have been made in recent years to study single particle breakage and the progeny fragment distribution (see, e.g., Schallnus and Schwedes, 1986). However the results are presented in terms of cumulative weight percentages, such as the  $B_{ij}$  values (see, e.g., Klimpel and Austin, 1984). Such results do not shed light on the manner in which a particle breaks, i.e., into the number of fragments and their sizes. It is proposed that particles, all formed in a similar manner will have similar fracture resistance, which has been confirmed (see, e.g., Schallnus and Schwedes, 1986). Then, by the application of similar kinds of stresses, these particles may break into similar number of fragments. Observations on the number of the daughter fragments of different sizes produced from the breakage of a single particle will yield these breakage probabilities. The  $\beta_{i,j}(k)$  values can be calculated from such measurements and knowledge of the rate of selection of particles for breakage, corresponding to  $S_i$  values reported in the literature (see, e.g., Shoji et al., 1980).

Attempts have been made to study the rate of size reduction for the three mechanisms in autogenous and semi-autogenous grinding by Austin et al. (1986), which is an important step towards understanding the internal dynamics of comminution operations. Similar studies can yield values for the abrasion and chipping rates,  $\alpha_j$  and  $\nu_j$ . The inclusion of

these mechanisms into the analysis offers a scope for scale-up as it is possible to predict the results of a larger operation if some measure of the kinds of stresses acting on individual particles together with a description of the internal mechanisms is known.

The numerical example describes the fluctuations that can be expected about the mean values of the sizes of the different particle groups. The covariances are useful in providing a qualitative picture of the evolution of the particle population. The standard deviation, which is the square root of the variance is roughly the order of the square root of the group size. Thus, stochastic modeling is a valuable tool to describe the inherent random processes prevailing in a comminution operation.

# NOTATION

$A_{i,k}$	=	first coefficient of the Fokker-Planck equation
$B_{i,j}$	=	second coefficient of the Fokker-Planck equation
$k$	=	state of the particle population
$N_i$	=	random variable representing the number of particles of a distinct feature in a particle population
$N_{i,j}$	=	random variable representing the number of particles of shape $i$ and volume $j\Delta V$ (or size $j$ ) in a particle population
$P(\{n\},t)$	=	joint probability distribution of random variables $\{N\}$
$p(\{n\},t)$	=	joint probability density function of random variables $\{N\}$
$s$	=	number of distinct size ranges into which the particle population is classified
$t$	=	time
$\Delta V$	=	volume of the smallest size; i.e., volume of the largest particle classified as a fine particle.
$W_t(\{n\}_0, \{n\}_1)$	=	transition intensity, transition rate or transition probability per unit time from state $\{n\}_0$ to $\{n\}_1$

## Greek Letters

$\alpha_j$	=	specific rate of abrasion of particles of size $j$ into those of size $j-1$ , time <sup>-1</sup>
$\beta_{i,j}(k)$	=	specific rate of breakage of size $i$ particles into two particles of sizes $j$ and $j-k$ , time <sup>-1</sup>
$\lambda_{i,j}$	=	rate of in-flow of particles of shape $i$ and size $j$ to the compartment, time <sup>-1</sup>
$\mu_j$	=	rate of exit of size $j$ particles from the compartment, time <sup>-1</sup>
$\langle \xi \rangle$	=	$\{n\}_1 - \{n\}_0$ : vector denoting the magnitude of the change in magnitude of random variables $\{N\}$ with elements $\xi_i$ or $\xi_{i,j}$

$\tau$  = time interval tending to zero

$v_j$  = specific rate of chipping of particles of size  $j$  into those of size  $j-1$ , time<sup>-1</sup>

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# APPENDIX A: CALCULATION OF COEFFICIENTS OF EQUATION (21)

The transition intensities are denoted by functions expressed as

$$W_t(\{n\}; \{\xi\}_k) = f_k(\{n\})$$

or

$$W_t(\{n\}; \underline{a}_k) = f_k(\{n\})$$

where  $\underline{a}$  is a vector in the space spanned by  $\underline{e}_1, \underline{e}_2, \dots, \underline{e}_r$ ;  $r$  = number of random variables.

$$\underline{e}_n = \begin{bmatrix} 0 \\ \cdot \\ \cdot \\ 0 \\ 1 \\ 0 \\ \cdot \\ \cdot \\ 0 \end{bmatrix} \quad n\text{-th row}$$

the elements of  $\underline{a}_k$  are equal to those of  $\{\xi\}_k$ , e.g., if

$$\{\xi\}_k = \{0, -1, 0, +1, +1\},$$

then

$$\underline{a}_k = \begin{bmatrix} 0 \\ -1 \\ 0 \\ +1 \\ +1 \end{bmatrix}$$

Let  $F$  be an  $n \times n$  matrix with off diagonal elements equal to zero and the diagonal element in column  $k$  equal to  $f_k(\{n\})$  for all  $k$ , i.e.,

$$F = [F(f_{kk} = f_k(\{n\}))]$$

where  $n$  equals the number of vectors,  $\underline{a}_1, \underline{a}_2, \dots, \underline{a}_n$ , i.e., the number of possible events.

Let  $W$  be a  $r \times n$  matrix with columns equal to the vectors  $\underline{a}_1, \underline{a}_2, \dots, \underline{a}_n$ , i.e.



$$W = [a_1, a_2, a_3, \dots, a_n]$$

Then, the matrix A of coefficients  $A_{i,k}(\{n\},t)$  is given by

$$A = WF$$

and the matrix B of coefficients  $B_{ij}(\{n\},t)$  is given by

$$B = WFW'$$

where  $W'$  is the transpose of  $W$ .

For instance, considering 3 random variables and 2 events, we have

$$W = \begin{bmatrix} a_{11} & a_{21} \\ a_{12} & a_{22} \\ a_{13} & a_{23} \end{bmatrix}$$

$$F = \begin{bmatrix} f_1 & 0 \\ 0 & f_2 \end{bmatrix}$$

$$A = \begin{bmatrix} a_{11} & a_{21} \\ a_{12} & a_{22} \\ a_{13} & a_{23} \end{bmatrix} \begin{bmatrix} f_1 & 0 \\ 0 & f_2 \end{bmatrix}$$

$$= \begin{bmatrix} a_{11}f_1 & a_{21}f_2 \\ a_{12}f_1 & a_{22}f_2 \\ a_{13}f_1 & a_{23}f_2 \end{bmatrix}$$

$$B = \begin{bmatrix} a_{11} & a_{21} \\ a_{12} & a_{22} \\ a_{13} & a_{23} \end{bmatrix} \begin{bmatrix} f_1 & 0 \\ 0 & f_2 \end{bmatrix} \begin{bmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \end{bmatrix}$$

$$= \begin{bmatrix} a_{11} & a_{21} \\ a_{12} & a_{22} \\ a_{13} & a_{23} \end{bmatrix} \begin{bmatrix} f_1 a_{11} & f_1 a_{12} & f_1 a_{13} \\ f_2 a_{21} & f_2 a_{22} & f_2 a_{23} \end{bmatrix}$$

$$= \begin{bmatrix} a_{11}f_1a_{11} & a_{21}f_2a_{21} & a_{11}f_1a_{12} & a_{21}f_2a_{22} & a_{11}f_1a_{13} & a_{21}f_2a_{23} \\ a_{12}f_1a_{11} & a_{22}f_2a_{21} & a_{12}f_1a_{12} & a_{22}f_2a_{22} & a_{12}f_1a_{13} & a_{22}f_2a_{23} \\ a_{13}f_1a_{11} & a_{23}f_2a_{21} & a_{13}f_1a_{12} & a_{23}f_2a_{22} & a_{13}f_1a_{13} & a_{23}f_2a_{23} \end{bmatrix}$$

These are the desired results. (Fox and Fan, 1986)

Table 1. Events, corresponding changes in magnitudes of the random variables, and associated transition intensities for the numerical example.

		Random variables representing particle populations of various shapes and sizes.				
		$N_{2,4}$	$N_{1,3}$	$N_{2,2}$	$N_{1,2}$	$N_{1,1}$
		Corresponding random variables with linearized subscripts				
		$N_1$	$N_2$	$N_3$	$N_4$	$N_5$
Event						
		$f_k = W_t \{ \{n\}; \{\xi\} \}$				
Abrasion	$\{\xi\}_1 = \{ 0, -1, 0, +1, -1 \}$	$f_1 = \alpha_3 n_{1,3}$				
	$\{\xi\}_2 = \{ 0, 0, 0, -1, +2 \}$	$f_2 = \alpha_2 n_{1,2}$				
Breakage	$\{\xi\}_3 = \{ -1, 0, +2, 0, 0 \}$	$f_3 = \beta_{2,4} (2) n_{2,4}$				
Chipping	$\{\xi\}_4 = \{ -1, +1, 0, 0, +1 \}$	$f_4 = u_4 n_{2,4}$				
	$\{\xi\}_5 = \{ 0, 0, -1, 0, +2 \}$	$f_5 = u_2 n_{2,2}$				
Inflow	$\{\xi\}_6 = \{ +1, 0, 0, 0, 0 \}$	$f_6 = \lambda_{2,4}$				
Outflow	$\{\xi\}_7 = \{ -1, 0, 0, 0, 0 \}$	$f_7 = \mu_{2,4} n_{2,4}$				
	$\{\xi\}_8 = \{ 0, -1, 0, 0, 0 \}$	$f_8 = \mu_{1,3} n_{1,3}$				
	$\{\xi\}_9 = \{ 0, 0, -1, 0, 0 \}$	$f_9 = \mu_{2,2} n_{2,2}$				
	$\{\xi\}_{10} = \{ 0, 0, 0, -1, 0 \}$	$f_{10} = \mu_{1,2} n_{1,2}$				
	$\{\xi\}_{11} = \{ 0, 0, 0, 0, -1 \}$	$f_{11} = \mu_{1,1} n_{1,1}$				

Table 2. Coefficients of the Fokker-Planck equation for the numerical example.

j	i				
	1	2	3	4	5
1					
$\lambda_{2,4}$	0	0	0	0	0
$-(\beta_{2,4} (2)^{u_4} \mu_{2,4}) N_{2,4}$					
$A_{1,j} =$					
2	$u_4 N_{2,4}$	$-(\alpha_3 + \mu_{1,3}) N_{1,3}$	0	0	0
3	$2\beta_{2,4} (2)^{N_{2,4}}$	0	$-(v_2 + \mu_{2,2}) N_{2,2}$	0	0
4	0	$\alpha_3 N_{1,3}$	0	$-(\alpha_2 + \mu_{1,2}) N_{2,2}$	0
5	$u_4 N_{2,4}$	$\alpha_3 N_{1,3}$	$2v_2 N_{2,2}$	$2\alpha_2 N_{1,2}$	$-\mu_{1,1} N_{1,1}$

Table 2. (cont.)

j	1	2	3	4	5
1					
1	$\lambda_{2,4}$	$-v N_{2,4}$	$-2\beta_{2,4} \{2\} N_{2,4}$	0	$-v N_{2,4}$
	$+\beta_{2,4} \{2\} + v_4 v_{2,4} N_{2,4}$				
B <sub>1,j</sub> = 2					
2	$-v N_{2,4}$	$-(\alpha_3 v_{1,3} N_{1,3} + v_4 n_{2,4})$	0	$-\alpha_3 N_{1,3}$	$-\alpha_3 N_{1,3} + v N_{4,2,4}$
3	$-2\beta_{2,4} N_{2,4}$	0	$4\beta_{2,4} \{2\} N_{2,4} + (v_2 v_{H,2}) N_{2,2}$	0	$-2v N_{2,2,2}$

Table 2. (cont.)

4	0	$-a_{3,1,3}^N$	0	$a_{3,1,3}^N$	$a_{3,1,3}^N$
				$+a_{2,1,2}^{*H,2}N_{1,2}$	$-2a_{2,1,2}^N$
5	$-v_{4,2,4}^N$	$v_{4,2,4}^N - a_{3,1,3}^N$	$-2v_{2,2,2}^N$	$a_{3,1,2}^N$	$v_{4,2,4}^N + a_{3,1,3}^N$
				$-2a_{2,1,2}^N$	$+4v_{2,2,2}^N + 4a_{2,1,2}^N$
					$+ \mu_{1,1,1}^N$

Table 3. Parameter values used in the numerical example.

$$\alpha_3 = 0.04 \text{ time}^{-1}$$

$$\alpha_2 = 0.03 \text{ time}^{-1}$$

$$\beta_{2,4}(2) = 0.08 \text{ time}^{-1}$$

$$u_4 = 0.05 \text{ time}^{-1}$$

$$u_2 = 0.04 \text{ time}^{-1}$$

$$\lambda_{2,4} = 200 \text{ time}^{-1}$$

$$\mu_{2,4} = 0.02 \text{ time}^{-1}$$

$$\mu_{1,3} = 0.02 \text{ time}^{-1}$$

$$\mu_{2,2} = 0.02 \text{ time}^{-1}$$

$$\mu_{1,2} = 0.02 \text{ time}^{-1}$$

$$\mu_{1,1} = 0.02 \text{ time}^{-1}$$

$$n_0 = 1000$$

# LIST OF FIGURES

- Fig. 1. Transient mean and variance of the population of size 4, regular particles in the compartment:

$$\square = \langle N_{2,4} \rangle, \Delta = \langle \langle N_{2,4}^2 \rangle \rangle.$$

- Fig. 2. Transient mean and variance of the population of size 3, regular particles in the compartment:

$$\square = \langle N_{1,3} \rangle, \Delta = \langle \langle N_{1,3}^2 \rangle \rangle.$$

- Fig. 3. Transient mean and variance of the population of size 2, irregular particles in the compartment:

$$\square = \langle N_{2,2} \rangle, \Delta = \langle \langle N_{2,2}^2 \rangle \rangle.$$

- Fig. 4. Transient mean and variance of the population of size 2, regular particles in the compartment:

$$\square = \langle N_{1,2} \rangle, \Delta = \langle \langle N_{1,2}^2 \rangle \rangle.$$

- Fig. 5. Transient mean and variance of the population of size 1, regular clusters of fines in the compartment:

$$\square = \langle N_{1,1} \rangle, \Delta = \langle \langle N_{1,1}^2 \rangle \rangle.$$

- Fig. 6. Correlation coefficients of the pairs of random variables  $\{N_{2,4}, N_{1,3}\}$ ,  $\{N_{2,4}, N_{2,2}\}$ ,  $\{N_{2,4}, N_{1,2}\}$  and  $\{N_{2,4}, N_{1,1}\}$ :

$$\square = \rho(N_{2,4}, N_{1,3}), \Delta = \rho(N_{2,4}, N_{2,2}), \\ * = \rho(N_{2,4}, N_{1,2}), \# = \rho(N_{2,4}, N_{1,1}).$$

- Fig. 7. Correlation coefficients of the pairs of random variables  $\{N_{1,3}, N_{2,2}\}$ ,  $\{N_{1,3}, N_{1,2}\}$  and  $\{N_{1,3}, N_{1,1}\}$ :

$$\square = \rho(N_{1,3}, N_{2,2}), \Delta = \rho(N_{1,3}, N_{1,2}), * = \rho(N_{1,3}, N_{1,1}).$$

- Fig. 8. Correlation coefficients of the pairs of random variables  $\{N_{2,2}, N_{1,2}\}$  and  $\{N_{2,2}, N_{1,1}\}$ :

$$\square = \rho(N_{2,2}, N_{1,2}), \Delta = \rho(N_{2,2}, N_{1,1}).$$

- Fig. 9. Correlation coefficient of the pair of random variables  $\{N_{1,2}, N_{1,1}\}$ :

$$\square = \rho(N_{1,2}, N_{1,1}).$$

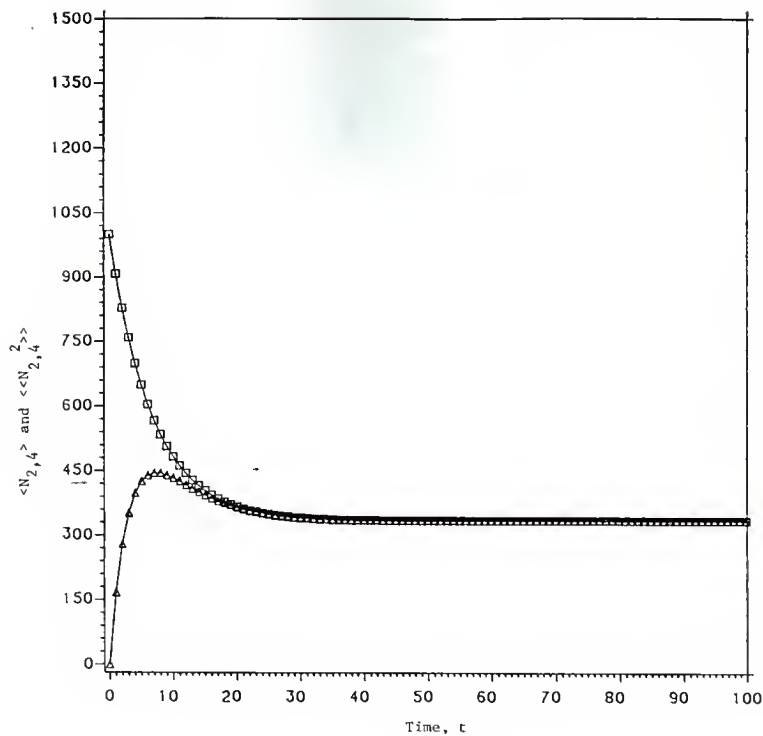


Fig. 1. Transient mean and variance of the population of size 4, regular particles in the compartment:

$$\square = \langle N_{2,4} \rangle, \Delta = \langle \langle N_{2,4}^2 \rangle \rangle.$$



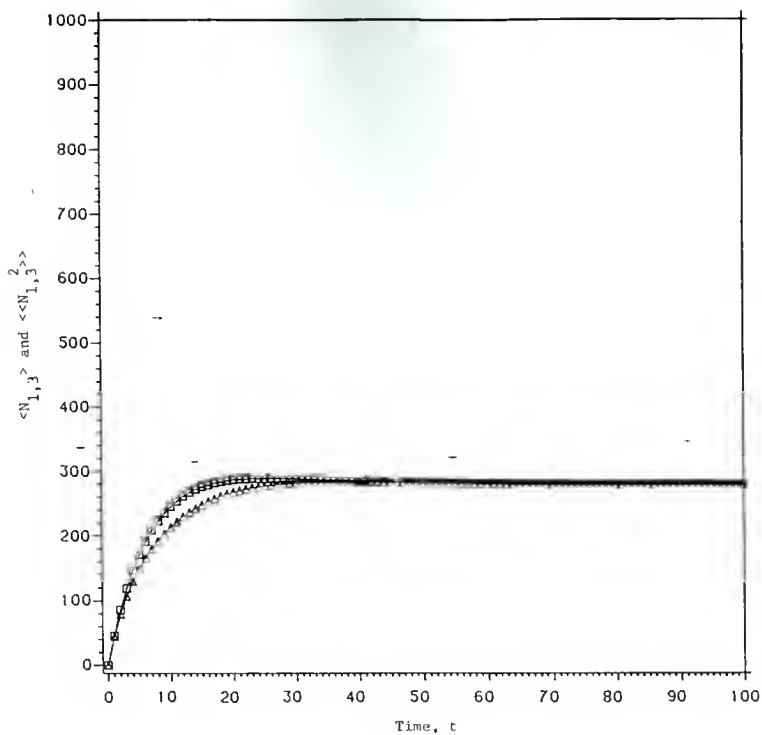


Fig. 2. Transient mean and variance of the population of size 3, regular particles in the compartment:

$$\square = \langle N_{1,3} \rangle, \Delta = \langle N_{1,3}^2 \rangle.$$

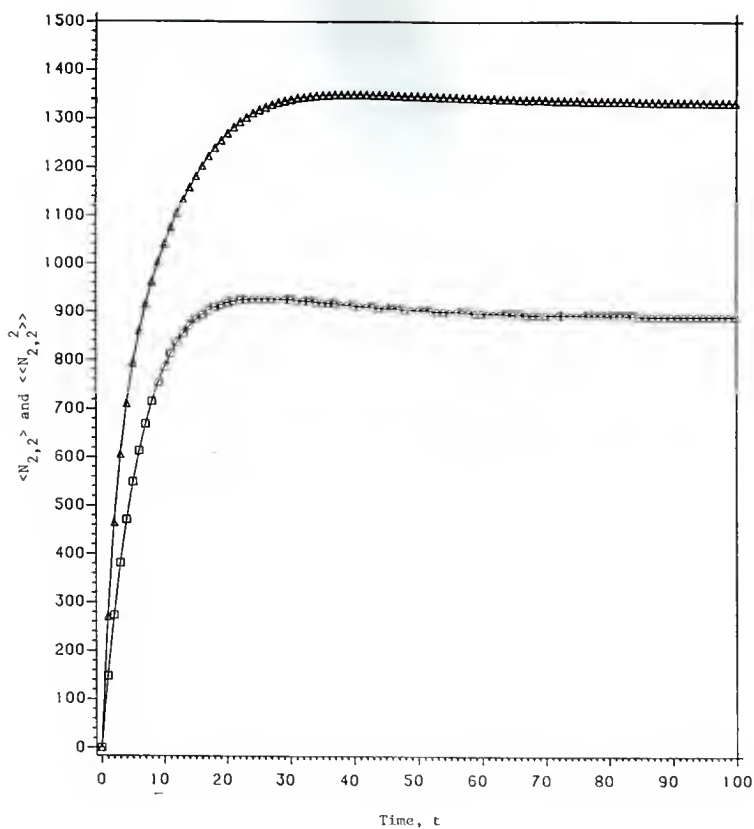


Fig. 3. Transient mean and variance of the population of size 2. Irregular particles in the compartment:

$$\square = \langle N_{2,2} \rangle, \Delta = \langle\langle N_{2,2}^2 \rangle\rangle.$$

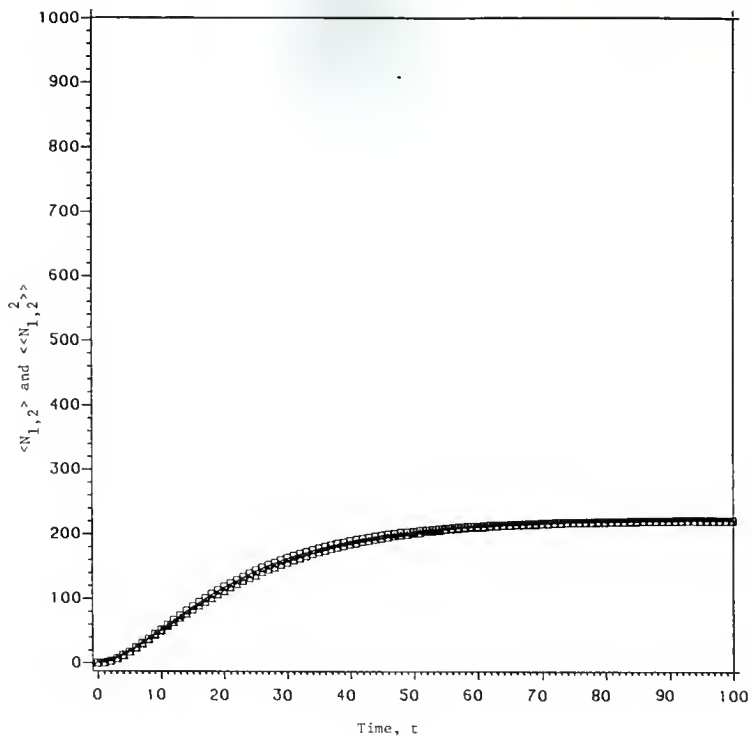


Fig. 4. Transient mean and variance of the population of size 2, regular particles in the compartment:

$$\square = \langle N_{1,2} \rangle, \Delta = \langle \langle N_{1,2}^2 \rangle \rangle.$$

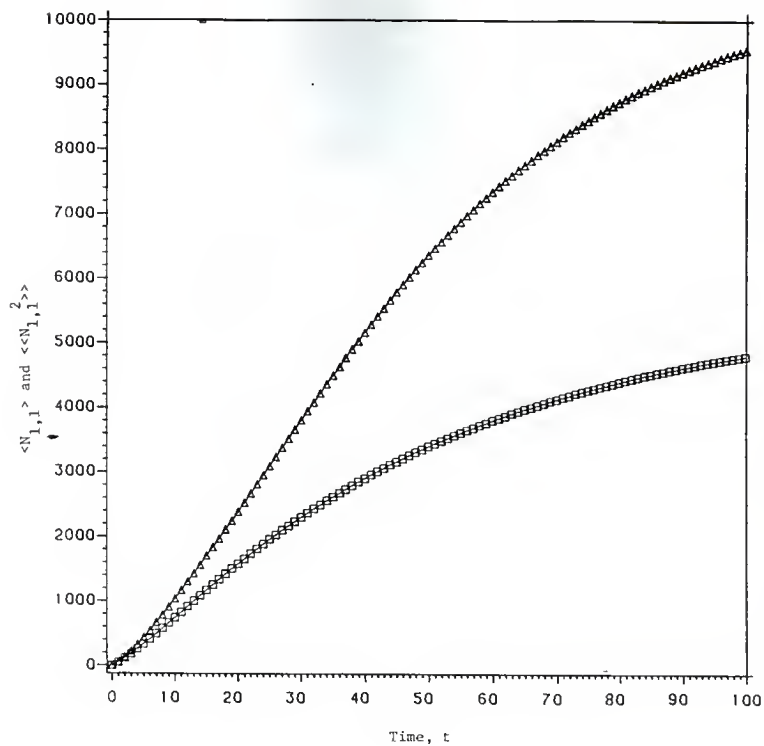


Fig. 5. Transient mean and variance of the population of size 1, regular clusters of fines in the compartment:

$$\square = \langle N_{1,1} \rangle, \quad \Delta = \langle\langle N_{1,1}^2 \rangle\rangle.$$

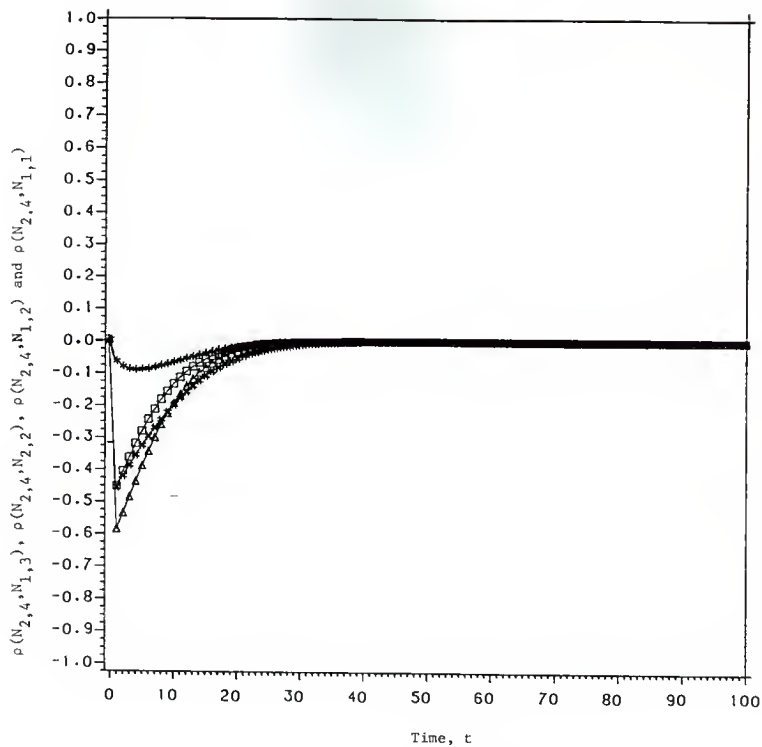


Fig. 6. Correlation coefficients of the pairs of random variables  $(N_{2,4}, N_{1,3})$ ,  $(N_{2,4}, N_{2,2})$ ,  $(N_{2,4}, N_{1,2})$  and  $(N_{2,4}, N_{1,1})$ :

$\square = \rho(N_{2,4}, N_{1,3})$ ,  $\Delta = \rho(N_{2,4}, N_{2,2})$ ,  
 $* = \rho(N_{2,4}, N_{1,2})$ ,  $\circ = \rho(N_{2,4}, N_{1,1})$ .

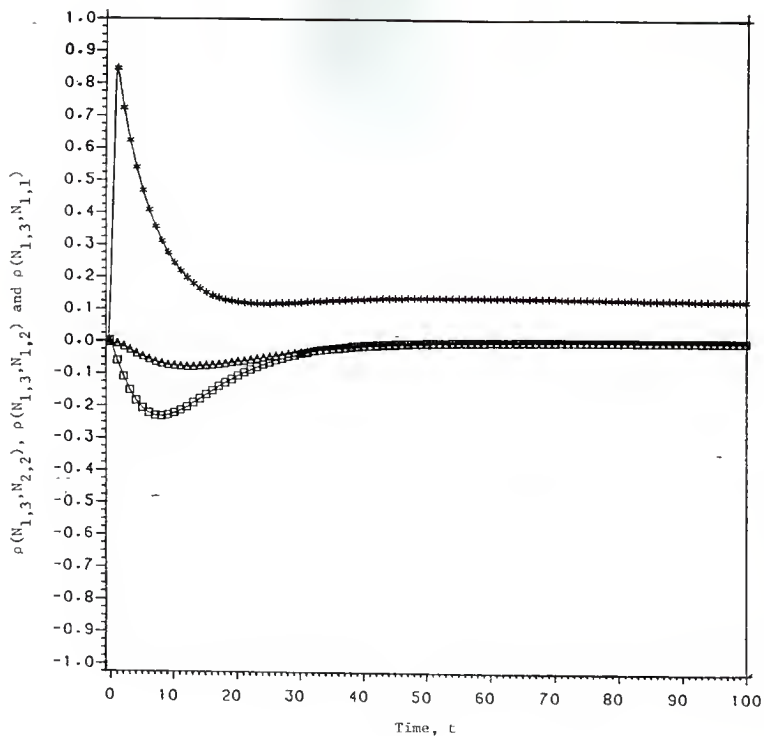


Fig. 7. Correlation coefficients of the pairs of random variables  $\{N_{1,3}, N_{2,2}\}$ ,  $\{N_{1,3}, N_{1,2}\}$  and  $\{N_{1,3}, N_{1,1}\}$ :

$$\square = \rho(N_{1,3}, N_{2,2}), \quad \Delta = \rho(N_{1,3}, N_{1,2}),$$

$$* = \rho(N_{1,3}, N_{1,1}).$$

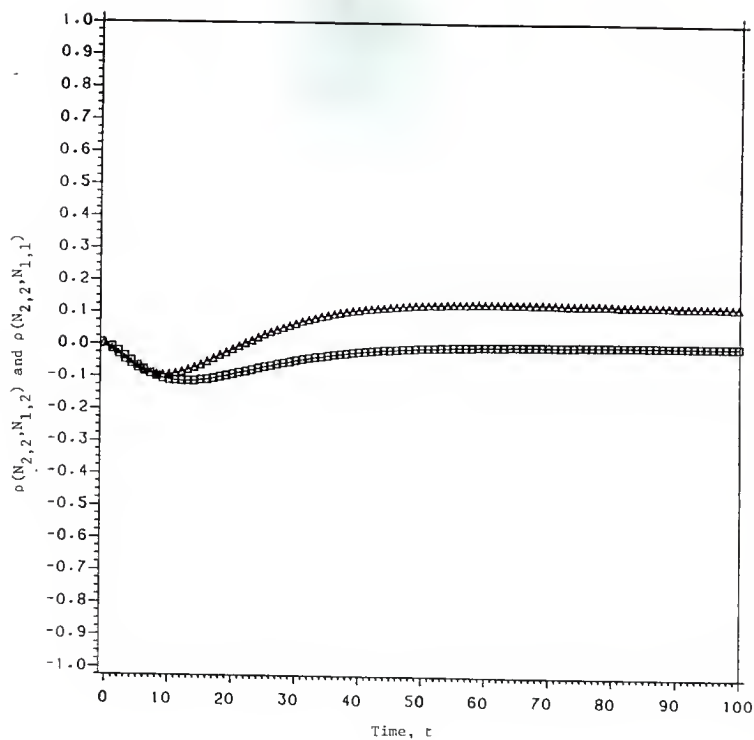


Fig. 8. Correlation coefficients of the pairs of random variables  $\{N_{2,2}, N_{1,2}\}$  and  $\{N_{2,2}, N_{1,1}\}$ :  
 $\square = \rho(N_{2,2}, N_{1,2})$ ,  $\Delta = \rho(N_{2,2}, N_{1,1})$ .

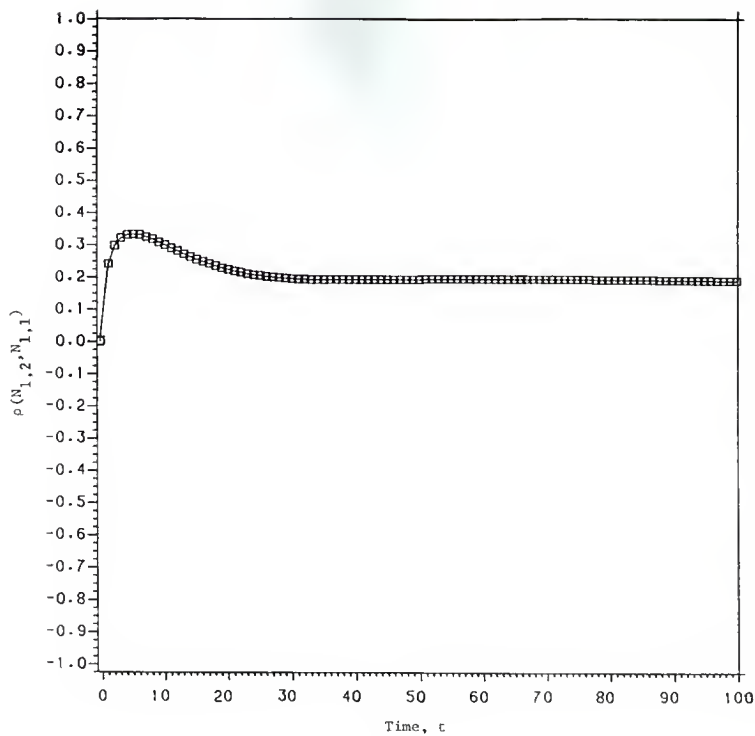


Fig. 9. Correlation coefficient of the pair of random variables  $\{N_{1,2}, N_{1,1}\}$ :

$$\square = \rho(N_{1,2}, N_{1,1}).$$



## CHAPTER 5

### CONCLUSIONS AND RECOMMENDATIONS

This study has yielded the following conclusions:

1. The probabilistic mesoscopic approach for describing a particulate system is based on stronger foundations than conventional macroscopic approaches, yielding hitherto unrecognized significant process characteristics. The stochastic formulations of system behavior and the establishment of the specific master equation in the present work provides an insight into the nature and mechanisms of a rate process without reference to deterministic dynamics, obtained as approximations to the probabilistic model. We can resort to such results to predict the results from operations at different scales.
2. The general cell model describing attrition behavior is modular and hence flexible. It can be applied to sections of a fluidized bed where different conditions exist such as the bed proper and freeboard zone.
3. The application of the general cell model to a fluidized bed predicts large fluctuations in the amount of fines entrained. This has an impact on elutriation studies as these fluctuations must be recognized and accounted for while obtaining data from experimental observations.
4. The master equation formulation has been demonstrated to be an effective mesoscopic approach in describing the particulate processes of sieving and grinding. An approximate solution has been

obtained in the case of non-linear sieving kinetics by using the system size expansion. Exact analytical solutions to the master equation have been obtained for the case of sieving without oversize particles. The master equation for the grinding process has been successfully approximated with a Fokker-Planck equation to yield the desired results.

5. It is necessary to distinguish between fluctuations from an inherently random process and those arising from the measurement techniques employed. Sieving, a widely employed particle classification method for particulate matter, is itself a random process as revealed through the study on sieving kinetics. Thus, the measured values of the product of attrition or grinding after sieving reflect fluctuations from two sources, namely, attrition or grinding and sieving.. This fact has not been recognized previously.
6. The results of the study on non-linear sieving kinetics indicate that the rate controlling step is the passage of the near mesh size particles. However, the definition of the near mesh size varies from material to material. This problem can be circumvented by introducing a probability associated with the initial near mesh size population on the sieve.
7. The stochastic analysis of comminution, based on the mesoscopic approach at the particle level with the inclusion of the different grinding mechanisms has yielded an improved treatment of the internal dynamics of the comminution operation over existing

process descriptions. Experimental techniques to measure the various rates are also proposed.

8. The concept of clusters of fines has been introduced in this work. The effect of fines on the system behavior is caused by these clusters, which can be measured more easily than individual fine particles. This is true in many cases such as particle entrainment from the fluidized bed.

Recommendations for future work are listed below.

1. Mechanical attrition in fluidized beds. In the absence of chemical attrition, inter-particle and particle-wall collisions cause attrition of the bed material. A master equation can be formulated for this problem. The transition intensities for inter-particle collisions will be non-linear and of second order. It is expected that inter-particle collisions predominate in wide beds and particle-wall collisions predominate in narrow beds. The relative values of these rates will determine the nature of the end product. At present no experimental evidence exists, indicating the second order breakage kinetics. This may be due to the fact that a large number of these experiments are conducted in narrow laboratory size fluidized beds.

2. Experimental studies on sieving operations. In the light of the theoretical studies on sieving kinetics, experimental observations providing the information required for the model's application

are necessary to determine the transition intensities and verify the model systematically.

3. Study of semi-batch and continuous sieving operations. The study of sieving kinetics can be extended to include semi-batch or continuous flow conditions. An interesting problem is that of studying the sieving kinetics in the case of a system of sieves placed one above the other in order of larger mesh size. Such systems are commonly used.

4. Presence of chipping during sieving operations. It has been observed that the material being sieved undergoes chipping randomly, i.e., the irregular feed which may be slightly oversize becomes near mesh size or near mesh size becomes slightly undersize and passes through the sieve. A stochastic analysis will provide information on the optimal sieving time or 'end point' of the sieving operation.

5. Experimental investigations on breakage probabilities of single particles. The theoretical work has indicated that the complex internal fracture mechanisms can be studied on a more feasible, particle level in terms of probabilities of breakage. It is necessary to know the number-size distribution of the progeny particles to determine the mode of breakage and form of the transition intensities. Existing experimental results indicate the cumulative size distributions of progeny particles which is inadequate for stochastic modeling. A slight modification in the design of such experiments would yield desired results.

6. Monte-Carlo simulation of particle size reduction. In order to obtain a deeper insight into the rate of selection for breakage and the subsequent mode of breakage, it may be necessary to simulate the grinding operation using techniques such as the Monte-Carlo method. This numerical technique is also useful in solving problems for which the system size expansion is invalid.

7. Experimental study on grinding mechanisms. Tracer methods can be utilized to determine the transition intensities of abrasion, breakage and chipping grinding mechanisms. Further study is required to design these experiments.

8. Stochastic analysis of autogeneous grinding. In autogeneous grinding, particle breakage occurs through inter-particle collisions, where the raw feed itself serves as the grinding media. It is expected that breakage is more likely to occur when large particles of similar sizes collide in which case the transition rate of breakage will depend on the number of particles in the two groups to which these colliding particles belong. This is essentially non-linear grinding kinetics. Nevertheless, this may not be evident if the abrasion and chipping rates are much higher. Further theoretical and experimental studies are required to shed light on such processes.

STOCHASTIC ANALYSIS OF PARTICULATE PROCESSES - A STUDY OF ATTRITION,  
SIEVING AND GRINDING

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AN ABSTRACT OF A MASTER'S THESIS

submitted in partial fulfillment of the

requirements for the degree

MASTER OF SCIENCE

Department of Chemical Engineering

KANSAS STATE UNIVERSITY  
Manhattan, Kansas

1986

## ABSTRACT

Particulate processes or systems lend themselves to a stochastic analysis due to the finite number of entities present in volume elements, random behavior of the individual particles and the complexities encountered in attempts to analyze the process on a microscopic scale. In this study, a mesoscopic approach is adopted to study attrition, and sieving and grinding operations. The attrition behavior in a continuous flow particulate system is described by a model based on the well-known Migration and Illness-Death stochastic processes. The master equation resulting from a stochastic population balance is formulated for the sieving and grinding processes. A rational approximation procedure, the system size expansion, is employed to solve the master equation when it is not amenable to an explicit analytical solution. The application of the model is illustrated with a numerical simulation in all three cases. The stochastic description is highly useful in understanding and predicting the results of processes involving finite entities and characterized by complex, obscure behavior on the microscopic scale, and providing a measure of the inherent fluctuations. This yields information on process characteristics, essential for understanding and developing the process under consideration.